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ZERO-GRAVITY ATMOSPHERIC CLOUD PHYSICS  
EXPERIMENT LABORATORY-  
ENGINEERING CONCEPTS/DESIGN TRADEOFFS

Volume II: Detailed Approaches

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16. ABSTRACT  This study summarizes work accomplished from January 1974 to October 1974 for the Zero-Gravity Atmospheric Cloud Physics Laboratory. This project involves the definition and development of an atmospheric cloud physics laboratory and the selection and delineation of a set of candidate experiments that require the unique environment of zero gravity or near zero gravity.  The primary goal of this effort is to define the experiment program and the laboratory concept for a Spacelab payload to perform cloud microphysics research. This multimission laboratory is to be available to the entire scientific community to utilize in furthering the basic understanding of cloud microphysical processes and phenomenon, thereby contributing to improved weather prediction and ultimately to provide beneficial weather control and modification.  * Volume I: Study Results, is available as CR-120500					
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## FOREWORD

The results reported herein encompass study efforts performed on the "Zero-Gravity Atmospheric Cloud Physics Experiment Laboratory - Engineering Concepts/Requirements/Design Trade-Offs Study" conducted for the NASA Marshall Space Flight Center (MSFC) by the McDonnell Douglas Astronautics Company. This report supplements and updates the information provided by the "Zero-Gravity Cloud Physics Laboratory Candidates Experiments Definition and Preliminary Concept Study" and contained in NASA reports CR 128998, CR 129002, and CR 129013.

The primary goal of the above efforts is to define the experiment program and the laboratory concept for a Spacelab payload to perform cloud micro-physics research. This multimission laboratory is to be available to the entire scientific community to utilize in furthering the basic understanding of cloud microphysical processes and phenomenon, thereby contributing to improved weather prediction and ultimately to provide beneficial weather control and modification.

The study scope performed by the McDonnell Douglas Astronautics Company, Biotechnology and Space Sciences Subdivision, involved the following tasks:

### Task 1 - Experiment Laboratory Subsystem Requirements

Experiment laboratory subsystem requirements were reviewed, expanded, and finalized in accordance with the technical guidelines. The experiment classes were utilized to establish the engineering design requirements for the cloud chamber's subsystems. The cloud chamber's subsystems and the experiment classes were evaluated to establish the parameter ranges and tolerances for the ancillary subsystems, as well as for data recording, transmission, and storage equipment. Experiment classes were used to determine the laboratory geometry requirements and operational controls.

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#### Task 2 - Experiment Laboratory Subsystem Definition

Tradeoff evaluations were conducted on all the cloud chambers and ancillary subsystems to establish final subsystem definition. Each subsystem was defined to the component level, and subsystem schematics were prepared. Tradeoffs were based on standard engineering criteria (weight, power, volume, development, status, etc.), Spacelab payload requirements, and with regard to keeping development costs and schedule increases at a minimum.

#### Task 3 - Experiment Laboratory Technology/Development

Component functional requirements and design specifications were developed in sufficient depth to permit evaluation of currently existing component technology. A survey of component status was conducted and a detailed status summary prepared. This status list identified (1) components currently existing that meet laboratory requirements, (2) components that can be readily modified, and (3) technology-deficient components and equipment that will require prototype development. Component development plans were formulated, in conformance with the subsystem definitions, for all technology deficient components. Subsystems contamination specifications must be established for the various experiment hardware configurations. Procedures must be established for meeting the contamination specifications and maintaining the required cleanliness levels throughout the mission.

#### Task 4 - Subsystems Evaluation

##### Cloud Chambers

Assessment of selected cloud chamber subsystem concepts requiring technology advancement or design modification was conducted. The effort performed placed primary emphasis on chamber compatibility with envisioned experiment usage on Spacelab. Priority was given to cloud chamber subsystems required for the "high-priority" experiments that have identified technology deficient components. Cloud chamber subsystems were evaluated to provide assurance of both compatibility with the envisioned experiment classes and design adequacy for experimenter-astronaut operation, including safety considerations.



### **Ancillary Equipment**

Selected ancillary subsystems requiring technology advancement or design modification were evaluated. The effort performed placed primary emphasis on subsystem compatibility with envisioned experiment usage on a Spacelab. Ancillary subsystems were evaluated to provide assurance of design adequacy over the required parameter ranges and tolerances, with both manual and automatic operational control. Evaluation of safety factors and maintenance concepts was performed.

### **Task 5 - Laboratory Definition**

The definition of the experiment laboratory was performed to the level required to permit fabrication of a soft mockup. The laboratory definition reflected the results of the program analysis conducted under this scope of work. The laboratory was based on updated Space Shuttle and Spacelab capabilities, and upon criteria in Appendix A and Appendix B in the RFP. Subsystem schematics reflecting the results of program experimentation were prepared and defined equipment to the component level. Interfaces between subsystems and between the cloud physics laboratory and the Spacelab were delineated. Location of subsystems and equipment within the laboratory were specified. Estimates of weight, power, volume, development, fabrication, and operation costs, mission timeline, and development schedule were provided.

### **Task 6 - Data Management**

A preliminary plan for managing data and its processing requirements was accomplished to include a conceptual operational mode for the laboratory based on current Shuttle system concepts. A data management and processing plan was provided that shows the relationship with raw data, filtered data, data handling techniques, etc., and all interfaces as required relative to ultimate delivery of data to the experimenter. Typical flow patterns were developed.

### **Task 7 - Cost, Schedule, SRT Requirement**

The contractor identified and defined (Phase B - C/D) the costs and cost spreads required to design, develop, and fabricate the laboratory equipment, instruments, instrumentation, etc. The contractor identified a development

schedule for the laboratory (Phase B - C/D) and also identified the required areas of supporting research and technology (SRT) required to support the development of the cloud physics laboratory. All priority categories of SRT technology were included as part of the basic laboratory development efforts by the contractor or his subcontractors.

This project is being conducted on behalf of NASA's Office of Application and Office of Manned Space Flight. The progress on this Space Shuttle/Spacelab payload has been enhanced by the enthusiastic response and support provided by the members of the cloud physics scientific community. This support is based on the recognition of the significant potential that such a payload may provide to the advancement of the basic understanding of cloud microphysical processes and phenomena. Comments on the contents of this report will be welcomed.

## INTRODUCTION

Understanding the microphenomena of cloud physics is one prerequisite to accurate prediction and practical modification of the weather. This understanding can be furthered by conducting certain experiments outside the influence of gravity — hence NASA's Cloud Physics Laboratory, scheduled to begin operating with Spacelab aboard the Shuttle in the early 1980's.

Why does one cloud produce a torrent of large, hammering raindrops — a cloudburst — while others precepitate only a fine drizzle? Why does one cloud rapidly develop and separate electrical charges, producing lightning and thunder, while other, outwardly similar clouds do not? Out of a field of thousands of clouds, several hundred of which contain thunderstorms, why does one develop into a tornado?

It has long been recognized that the answers to these and many other questions about atmospheric behavior depend to a substantial degree upon obtaining a better understanding of the microphenomena of cloud physics. For the past 30 years microphysicists, physical chemists, applied physicists, and meteorologists have been deeply involved in concentrated studies aimed at developing such an understanding. Their work — combined with advances in field observation, data gathering, and information processing — has led to the improved forecasting we enjoy today, as well as to our limited capability to stimulate or inhibit precipitation. In fact, the National Advisory Committee on Oceans and Atmospheres (NACOA) states in its Second Annual Report to the President and the Congress that ". . . we appear to stand on the threshold of practical weather modification. . . ."

The ability to control the weather can bring enormous benefits to man. It has been estimated that the ability to forecast accurately for only five days in advance would make possible savings of \$2-1/2 billion per year in agriculture, \$3 billion in management of water resources, \$75 million in retail marketing, and \$100 million in surface transportation.

These estimates are for the United States alone. Worldwide figures would be many times as great. And if such savings could result from the ability to forecast precisely and reliably for a mere five days ahead, the savings and increased productivity that would arise from the ability to predict the weather for months in advance, and to change it materially in desired localities, would be almost unimaginable.

A great deal of research remains to be done before weather forecasting and control capabilities of such scope can be achieved. NACOA points out that ". . . it is now time to increase the relative effort on smaller-scale meteorological phenomena . . .", while the joint Panel on Weather and Climate Modification of the National Academy of Sciences and the National Research Council has underscored the need for quantitative information at the micro-phenomenal level. Processes such as nucleation, growth of droplets and ice crystals, generation and separation of electrical charges, and "scavenging" (collection of gases and other atmospheric constituents by droplets and crystals) are of fundamental importance. Many aspects of these micro-phenomena are independent of gravity, and adequate observation under laboratory conditions demands that gravitational effects be absent.

It is the mission of NASA's Cloud Physics Laboratory (CPL) to make these gravity-free observations practical, productive, and economically feasible. Conceived as a reusable, general-purpose facility for manned research in low earth orbit, the CPL will be available for experiments originating both outside and within the government. With a planned operational life of several years, the laboratory will accommodate a broad range of experiments and will be adaptable to the program dynamics and the needs for supplementary experiments that will develop as the investigations progress.

The laboratory will be flown aboard the Space Shuttle (Figure 1) as a "partial Spacelab payload. (The CPL will use only a portion of the bay space, subsystem support, and mission time available, and thus will be combined with other payloads to make up a Shuttle/Spacelab mission.) The astronaut-experimenter, working in a shirtsleeve environment, will conduct the cloud physics experiments under the direction — via real-time communication,



Figure 1. Space Shuttle Rendering

where necessary — of principal investigators on the ground. First operations will be in the early 1980's.

#### GRAVITY: THE NEED FOR ITS ABSENCE

The difficulties that gravity creates in cloud physics investigations are related to the physical dimensions of the field in which the phenomena of interest occur in nature, as compared to the size of the observational volume that can be constructed in the laboratory.

A large cloud may be many cubic miles in volume. The constituents within the cloud — nuclei, droplets, crystals, gases, and so on — are free to move for miles in buoyancy or free fall. During these movements, they are constantly forming, dissipating, associating, disassociating, or passing from one state of matter to another. The average lifetime of discrete particles or groups of particles is 20 to 30 minutes. Within this time, exchanges of electricity and heat take place; droplets and crystals grow, freeze, thaw, evaporate, or precipitate; and the redistribution of energy that produces our weather occurs. Particle diameters are extremely small (0.1 micron is common), and differences in temperature and electrical charge, though often minute, are highly critical.

The researcher attempting to study these phenomena in the terrestrial laboratory must work with a pressure-temperature-humidity chamber that encloses only a minuscule fraction of the volume contained in a natural cloud. When with elaborate care he has produced within this chamber the particles that he wishes to study, he has only a few seconds to observe them before they drift to the chamber floor. If he requires a longer time, he must somehow hold his specimen in place against the pull of gravity. Techniques used for this purpose have included capturing individual particles and placing them on surfaces of waxed paper, teflon, copper, or stainless steel; hanging them on thermocouples; suspending them on threads or on fragments of a spider's web; and "clamping" them between the surfaces of two immiscible liquids. All of these methods have been used with some success — and all have been of only limited value because they introduce effects of forces

greater in magnitude than those being studied, or because they prevent interactions among atmospheric constituents.

Several approaches have been tried in attempts to avoid gravitational effects for longer periods of time. Drop towers have been used to obtain near-zero-g conditions for up to four seconds, and aircraft flying on low-g parabolic trajectories can permit observations 14 to 20 seconds long. Vertical wind tunnels, by suspending particles in an upward-moving column of air, have permitted the 20- to 30-minute experiment durations required, but only a small range of particle diameters can be observed at one time because particles of different sizes differ in aerodynamic buoyancy and uniformity of particle size does not occur in nature.

Sounding rockets and automated satellites have been considered for cloud physics research, but neither has been used. Sounding rockets have been neglected because no more than 10 minutes of experiment time could be obtained per flight, and satellites have had no serious advocates because only limited power, volume, weight, and expendable supplies could be put into orbit — at great expense for experiment automation, data management, thermal and attitude control, and launch.

The manned orbital approach, in contrast, entails no significant constraints. Five or more days of experiment time will be available on Shuttle/Spacelab missions. Particles can be floated in the cloud chamber for the natural duration of the processes being observed. Conditions of temperature, pressure, and illumination can be varied as needed, while thermal, electrical, and moisture parameters can be closely controlled and easily altered. Thus particles can be grown, diminished, frozen, thawed, evaporated, and allowed to migrate, collect, combine, and dissociate just as they would do in a natural cloud — all under conditions conducive to microscopic observation, photography, and precision instrumentation.

By providing these capabilities, the Cloud Physics Laboratory will make possible investigations that could not be conducted before — but it will by no means supersede the terrestrial laboratory. As indicated in Figure 2, many cloud

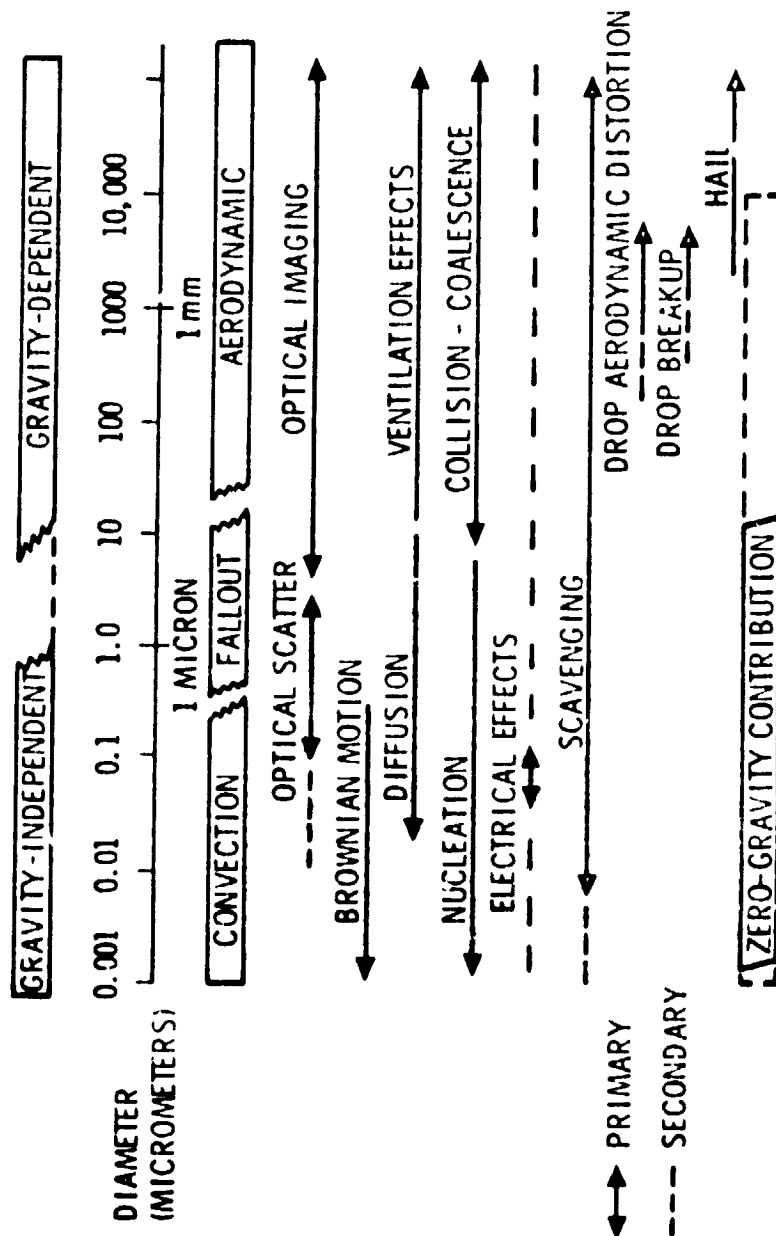


Figure 2. Cloud Physics Regime



microphenomena are gravity-dependent or involve gravity-dependent factors, and much work remains to be done in these areas.

Nor will the CPL go aloft without owing a direct debt to earlier low-g techniques. Drop towers and parabolic aircraft flights have application for development of laboratory elements such as droplet generators and environment controllers.

As a complement to terrestrial laboratories, the CPL may produce technological-fallout benefits for earth-based cloud physics research. At present, terrestrial laboratories tend to be highly specialized for investigation of closely defined microphysical processes. Hence they usually employ one-of-a-kind experiment chambers, instrumentation, and support systems. The CPL, which will use interchangeable chambers supported by standardized systems and a considerable degree of automation, will make available many efficient apparatus concepts and standardized designs that may both ease the tasks of experiment conception and preparation and furnish a basis for direct exchange and comparison of data among researchers.

#### THE CPL CONCEPT AND HOW IT GREW

The concept of a manned orbital laboratory for zero-g research in cloud physics was first systematically evaluated during the late 1960's as part of an analysis of research and engineering requirements in oceanography and meteorology. This analysis, sponsored by Marshall Space Flight Center, found that the laboratory would be prohibitively expensive if it were self-contained and independently launched. However, it would be economically attractive if it could be attached to and supported by a host vehicle that would provide environmental control, life support, attitude control, data management, communications, and crew quarters for common support of several similar laboratories. Such a host vehicle, the Space Station, was then under study and appeared able to accommodate the CPL with minimum impact on operations, weight, volume, and support systems.

Discussions followed between MSFC and leading cloud physicists in universities, government laboratories, and private research organizations. These

contacts led to the establishment of the Cloud Physics Laboratory as a NASA/MSFC project sponsored by the Office of Applications.

The project approach was aimed from the outset at responding to the research needs of the scientific community. Experiment suggestions were solicited from more than 200 scientists representing 54 organizations, and over 20 visits were made to organizations and laboratories. The experiment suggestions submitted in response, and the corresponding equipment requirements, formed the basis for development of experiment classes and preliminary definition of the laboratory design.

Individual scientists participated in organizing the experiment suggestions into classes and in defining objectives, approaches, experimental methods, equipment, instrumentation, and data to be obtained for each class. The results of these efforts, together with the laboratory concept that evolved in parallel with the classification and survey work, were reviewed by a group of distinguished cloud physics researchers.

This same approach has been used to refine and adapt the experiment classes as planning for the national space effort has progressed from the original Space Station concept to the current Shuttle/Spacelab combination. The present experiment classes are summarized. The requirements of these 21 classes can be met by six cloud-chamber types. Two or more Shuttle/Spacelab flights per year will be needed over a 10-year period.

The laboratory (Figures 3 and 4) will occupy a portion of a Spacelab module mounted in the Shuttle Orbiter payload bay and will be dependent upon the host configuration for power, heat rejection, data management, communications, and astronaut-experimenter accommodations. The CPL console as currently defined will be 2.70 meters in length, 2.72 meters in height, and 1.24 meters in maximum depth. Laboratory weight will be between 998 and 1368 kilograms and average power demands will range from 756 to 1155 watts.

The primary criteria underlying laboratory design definition have been value to the experiment program, low cost, and flexibility to accommodate evolving program requirements. Basic subsystems will be designed to remain

# SUMMARY OF EXPERIMENT CLASSES

Experiment Class Number and Title	Primary Chamber*	Alternate Chamber*
1. Condensation Nucleation	CFD	E
2. Ice Nucleation	SDI	E
3. Ice Multiplication	SDI	E
4. Charge Separation	SDI	G
5. Ice-Crystal Growth Habits	SDI	E
6. Scavenging	SDI	G
7. Riming and Aggregation	SDI	G
8. Droplet-Ice Cloud Interactions	SDI	E
9. Homogeneous Nucleation	SDI	E
10. Collision-Induced Freezing	SDI	G
11. Saturation Vapor Pressure	SDI	E
12. Adiabatic Cloud Expansion	E	-
13. Ice Nuclei Memory	E	SDI
14. Terrestrial Expansion Chamber Evaluation	E	-
15. Condensation Nuclei Memory	E	SDL
16. Nuclei Multiplication	G	E
17. Drop Collision Breakup	G	SDI
18. Coalescence Efficiencies	G	SDI
19. Static Diffusion Chamber Evaluation	SDL	-
20. Unventilated Droplet Diffusion Coefficients	SDL	E
21. Earth Simulation	ES	-

\*CFD = Continuous-flow diffusion

E = Expansion

G = General

SDL = Static diffusion, ice

SDL = Static diffusion, liquid

ES = Earth Simulation

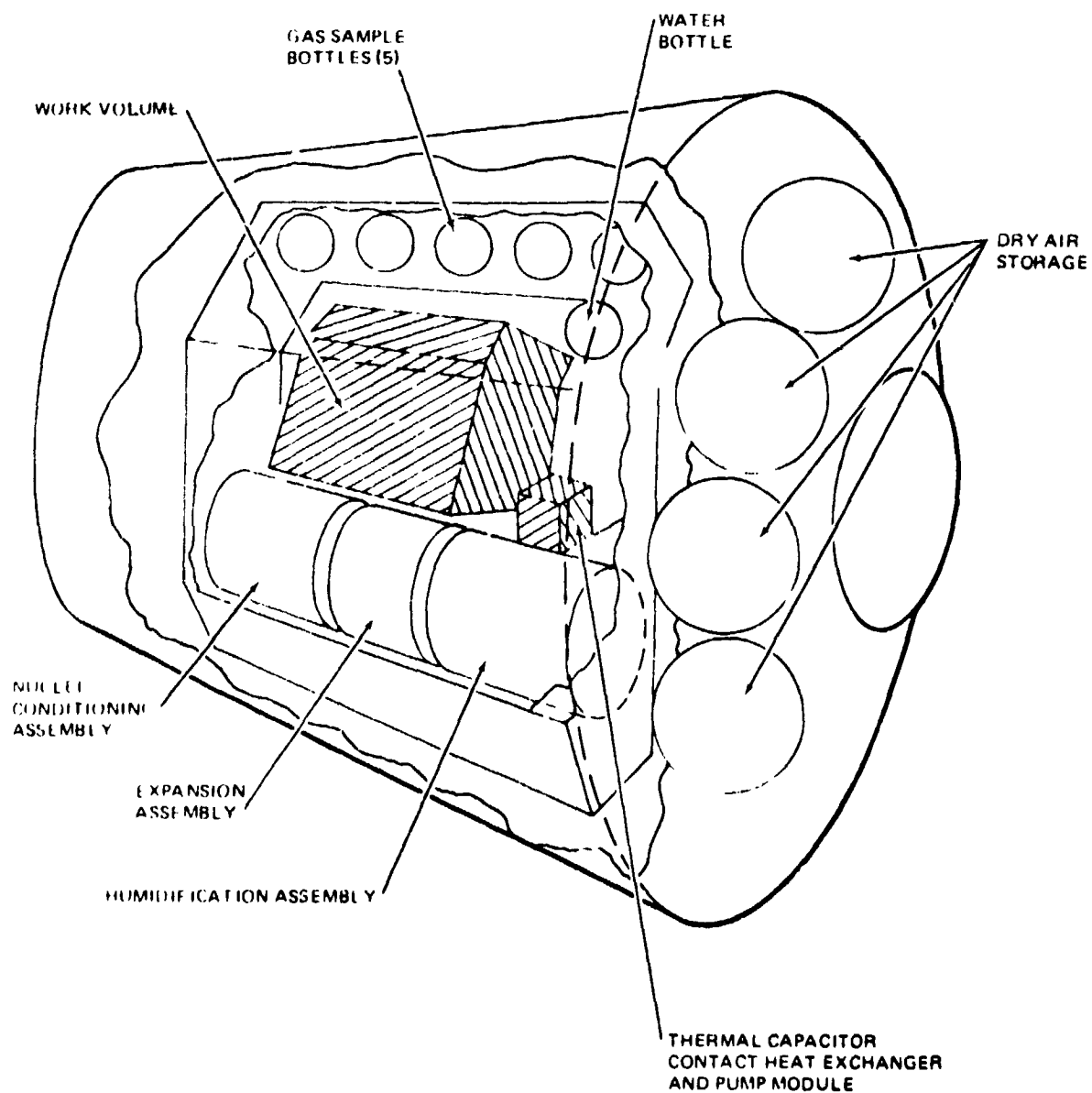


Figure 3. Cloud Physics Laboratory -- Outboard View

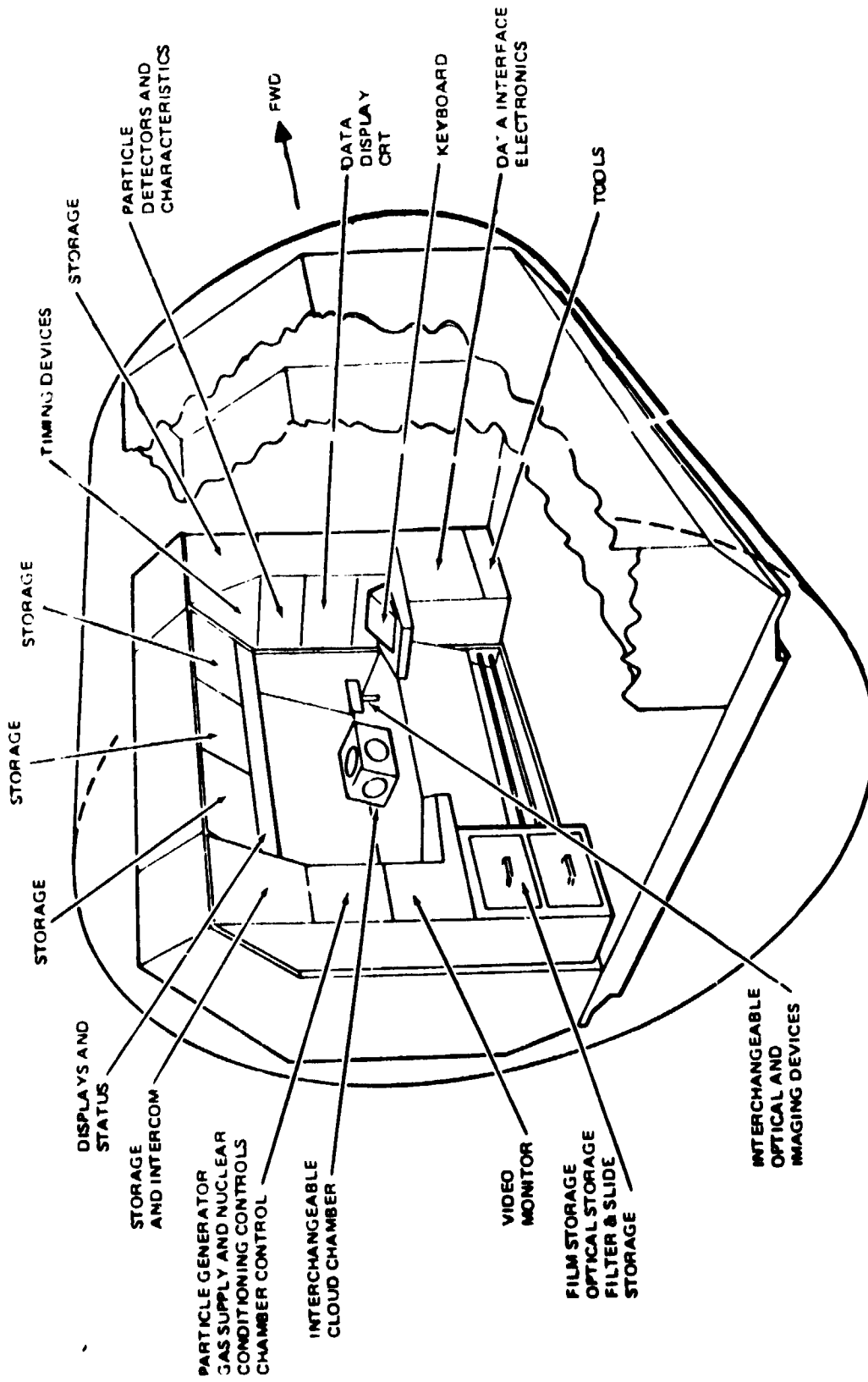


Figure 4. Cloud Physics Laboratory - Inboard View

the same from mission to mission, but features permitting convenient and economical modification or changeover to advanced subsystems will be incorporated to provide for growth and contingencies during the projected operational lifetime. Instrumentation, control, and data recording will feature extensive automation with manual override.

Five standardized cloud chambers (Figure 5) will constitute the primary experiment apparatus. These chambers will be fundamentally identical to those used in terrestrial laboratories but will be distinguished by design features tailored to manned orbital experimentation. Among these features will be heat pipes for thermal control (to conserve power), reduced thermal mass (to reduce weight and increase efficiency in use of experiment time), water reservoirs and flow controls designed for zero gravity, and provisions for meeting the stringent standards of safety and reliability essential to manned space operations. A macroscale experiment, using an earth simulation chamber (ES), has shown a high degree of commonality with microscale experimentation and is presently defined for inclusion in the CPL concept.

#### THE LABORATORY IN USE

CPL project planning is keyed to making the laboratory available on a schedule compatible with achievement of initial operational capability by the Shuttle and Spacelab. The first two laboratory launches from Kennedy Space Center will be separated by six months. The intervals between missions thereafter will be based on the hours of experimentation required, the time necessary to prepare for each mission, and the flight frequency needed to complete the experiment program within a reasonable period.

Two CPL's are envisioned at this time. Assessment of factors such as flight frequency, preparation time required between flights, design life, life-cycle cost, and flexibility of mission opportunities indicates that this number of units will be adequate to assure attainment of project objectives. Normally, one of the laboratories will be in launch preparation and in orbit while the other is being repaired, refurbished, and modified after completion of a mission. In coping with contingencies, the existence of two units — combined with designed-in economy and rapidity of repair and refurbishment—

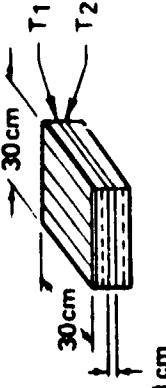
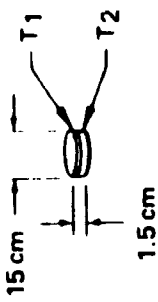
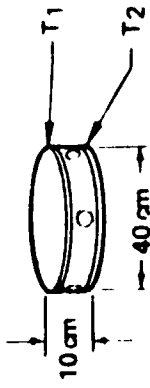
<p>PRESSURE RANGE TOLERANCE</p> <p>RELATIVE HUMIDITY TOLERANCE</p> <p>TEMPERATURE TOLERANCE</p> <p><math>\Delta T (T_1 - T_2)</math></p> <p>FLOW RATE</p>	 <p>CONTINUOUS - FLOW DIFFUSION (CFD)</p> <p><math>T_2 &lt; T_1</math> LIQUID SURFACES <math>0.01 \mu m &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 10 \mu m</math> OUTPUT - SIZE DISTRIBUTION CONDENSATION NUCLEATION STUDIES</p>	 <p>STATIC DIFFUSION, LIQUID (SDL)</p> <p><math>T_2 &lt; T_1</math> LIQUID SURFACES <math>0.01 \mu m &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 10 \mu m</math> OUTPUT - NUMBERS ONLY CONDENSATION NUCLEATION STUDIES</p>	 <p>STATIC DIFFUSION, ICE (SDI)</p> <p><math>T_2 &lt; T_1</math> ICE SURFACES <math>1 \mu m &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 1 \mu m</math> OUTPUT - SIZE, SHAPE ICE CRYSTAL STUDIES</p>	<p>760 TO 700 mm ABS <math>\pm 5</math> mm REL <math>\pm 1</math> mm</p> <p>100% TO 103% ABS <math>\pm 0.02\%</math> REL <math>\pm 0.01\%</math></p> <p>0°C TO +35°C ABS <math>\pm 0.1^\circ\text{C}</math> REL <math>\pm 0.02^\circ\text{C}</math></p> <p>0 TO 10°C</p> <p>0.25 SCFM</p>
	<p>760 TO 140 mm ABS <math>\pm 10</math> mm REL <math>\pm 1</math> mm</p> <p>100% TO 103% ABS <math>\pm 1\%</math> REL <math>\pm 0.5\%</math></p> <p>0°C TO +35°C ABS <math>\pm 0.1^\circ\text{C}</math> REL <math>\pm 0.02^\circ\text{C}</math></p> <p>0 TO 10°C</p>	<p>760 TO 140 mm ABS <math>\pm 10</math> mm REL <math>\pm 1</math> mm</p> <p>100% TO 103% ABS <math>\pm 1\%</math> REL <math>\pm 0.5\%</math></p> <p>0°C TO +35°C ABS <math>\pm 0.1^\circ\text{C}</math> REL <math>\pm 0.02^\circ\text{C}</math></p> <p>0 TO 10°C</p>	<p>760 TO 140 mm ABS <math>\pm 10</math> mm REL <math>\pm 1</math> mm</p> <p>80% TO 120% ABS <math>\pm 1\%</math> REL <math>\pm 0.5\%</math></p> <p>-60°C TO +35°C ABS <math>\pm 1^\circ\text{C}</math> REL <math>\pm 0.1^\circ\text{C}</math></p> <p>0 TO 60°C</p>	

Figure 5. Cloud Chambers (Sheet 1 of 2)

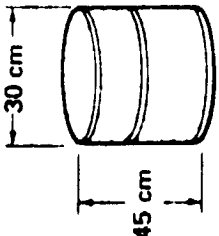
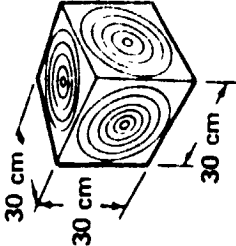
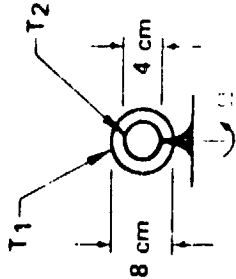
 <p>EXPANSION (E) HYDROPHOBIC SURFACES COOLED WALLS 0.01 <math>\mu\text{m}</math> &lt; PARTICLE DIAM. ETER &lt; 100 <math>\mu\text{m}</math> ADIABATIC EXPANSION OUTPUT - NUMBERS, MEAN SIZE CLOUD SIMULATION STUDIES</p>	 <p>GENERAL (G) ELECTRIC FIELDS DROPLETS &gt; 100 <math>\mu\text{m}</math> LARGE-PARTICLE INTERACTIONS</p>	 <p>EARTH SIMULATION (ES) DIFFERENTIALLY HEATED ROTATING SPHERICAL ANNULUS OF FLUID ROTATION RATE - 6 RAD/SEC PLANETARY AND SOLAR CONVECTION STUDIES</p>
<p>PRESSURE RANGE TOLERANCE</p>	<p>760 TO 140 mm ABS <math>\pm</math> 2 mm REL <math>\pm</math> 0.1 mm</p>	<p>AMBIENT N/A</p>
<p>RELATIVE HUMIDITY TOLERANCE</p>	<p>1 TO 100% ABS <math>\pm</math> 1% REL <math>\pm</math> 0.5%</p>	<p>N/A - -</p>
<p>TEMPERATURE TOLERANCE</p>	<p>-40°C TO +35°C ABS <math>\pm</math> 0.5°C REL <math>\pm</math> 0.05°C</p>	<p>+15°C TO +35°C ABS <math>\pm</math> 0.1°C REL <math>\pm</math> 0.03°C</p>
<p><math>\Delta T</math> (T<sub>1</sub> - T<sub>2</sub>)</p>	<p>-</p>	<p>0 TO 7°C</p>

Figure 5. Cloud Chambers (Sheet 2 of 2)



will permit the laboratories to serve as backups for each other and will impart an ability to take advantage of mission opportunities that may arise from failure of other payloads to meet their flight schedules. NASA, working closely with laboratory users, will be responsible for the CPL throughout the entire mission-preparation cycle, from refurbishment and modification through provisioning and mating with Spacelab and the Shuttle, to the launch itself.

It is expected that most laboratory users, particularly during the early portion of the operational phase, will represent the domestic and international scientific communities. For each mission, a team of principal investigators will be organized among the users, with selections and assignments being made by a panel of senior cloud physicists, meteorologists, cloud modelers, and field experimenters. The principal-investigator teams will formulate the detailed experiment objectives and timelines, assist in astronaut-experimenter training and launch preparations, support flight operations, debrief the astronauts, reduce and evaluate the experiment data, and prepare the experiment reports.

It is also anticipated that interests such as shipping, fisheries, forestry, air transport, and agriculture may wish to become involved in CPL operations. The participation of such potential users has so far been indirect (through contact with the scientific community), but as the project matures, the NASA procedure for announcement of flight opportunities will be used to alert special-interest organizations that may desire to support research, participate in mission planning, or engage directly in experimentation.

#### GETTING THERE FROM HERE

The Cloud Physics Laboratory project is now gathering the increased momentum that will culminate in orbital operations. Studies to refine the experiment program and laboratory concept were completed in 1973. Actual flight experience was gained on the Apollo 16, Apollo 17, and Skylab missions, during which precursor qualitative experiments and demonstrations of droplet dynamics substantiated the advantages of zero gravity for cloud

physics research and reiterated the value of man as an on-the-spot experimenter and decision-maker. In-depth definition studies now in progress will lead to preliminary design in 1975, followed by development and qualification extending from 1976 to 1980.

Much work remains to be done, but the benefits to be derived will make the journey from 1974 through the operational period well worth the traveling. Long-range prediction and control of the weather could prove to be one of the most important single development to evolve in the next few decades from space exploration. The Cloud Physics Laboratory is expected to be an important milestones in that evolution.

# CONTENTS

## VOLUME I

<b>Section 1</b>	<b>SUMMARY</b>	<b>1-1</b>
<b>Section 2</b>	<b>PROJECT STATUS AND PROGRESS</b>	<b>2-1</b>
2.1	Feasibility Study - First Phase (September 1971 to July 1972)	2-1
2.2	Feasibility Study - Second Phase (July 1972 to April 1973)	2-4
2.3	Feasibility Study - Third Phase (April 73 to September 1973)	2-7
<b>Section 3</b>	<b>EXPERIMENT PROGRAM SUMMARY</b>	<b>3-1</b>
3.1	Experiment Program	3-3
3.2	Cloud Chamber Selection	3-10
3.3	Research Related Equipment	3-16
3.4	Mission Assessment	3-16
3.5	Priority Mission Ranking	3-22
3.6	Priority Mission Set - Total Experiment Program Require- ments Comparison	3-24
<b>Section 4</b>	<b>CLOUD PHYSICS LABORATORY CONCEPT</b>	<b>4-1</b>
4.1	Laboratory Concept Rationale	4-1
4.2	Design Features	4-2
4.3	Guidelines and Constraints	4-3
4.3.1	Programmatics	4-3
4.3.2	Laboratory/Concept Justification	4-4
4.3.3	Operations	4-32
4.3.4	Laboratory Growth Potential	4-35
4.4	Scientific Community Participation	4-37
4.4.1	Project Participation	4-38
4.4.2	Selection Procedure (Tentative)	4-40

<b>Section 5</b>	<b>CLOUD PHYSICS LABORATORY SUBSYSTEMS</b>	<b>5-1</b>
<b>5.1</b>	<b>Thermal Control/Expendables Storage and Control</b>	<b>5-1</b>
5.1.1	Thermal Control	5-2
5.1.2	Flow, Humidity, and Pressure Control	5-5
5.1.3	Expendables Storage	5-5
5.1.4	Instrumentation and Display Assembly	5-8
5.1.5	Expendables	5-8
5.1.6	Cleansing Purge and Vent Assembly	5-9
<b>5.2</b>	<b>Particle Generator</b>	<b>5-9</b>
5.2.1	Wire Probe Retractor Generator	5-9
5.2.2	Water Drop Impeller Liquid/Ice Drop Generator	5-13
5.2.3	Vibrating Orifice Liquid/ Ice Droplet Generator	5-13
5.2.4	Evaporation/Condensation Aerosol Generator	5-15
5.2.5	Spray Atomization Nuclei Generator	5-15
5.2.6	Powder Dispersion Nuclei Generator	5-17
5.2.7	Particle Injector and Size Conditioner	5-17
<b>5.3</b>	<b>Data Management</b>	<b>5-19</b>
5.3.1	Control Processor Assembly	5-22
5.3.2	Tape Recorder Assembly	5-27
5.3.3	Master Control Assembly	5-27
5.3.4	Signal Conditioning Electronics Assembly	5-27
5.3.5	Instrumentation and Display Assemblies	5-28
5.3.6	Expendables	5-29
<b>5.4</b>	<b>Particle Detector and Characterizer</b>	<b>5-29</b>
5.4.1	Optical Particle Counters	5-30
5.4.2	Pulse Height Analyzer	5-35
5.4.3	Condensation Nucleus Counter	5-35
5.4.4	Microporous Filters	5-36
5.4.5	Piezoelectric Quartz Crystal Mass Monitor	5-36
5.4.6	Cascade Impactor	5-38
5.4.7	Electrical Aerosol Size Analyzer	5-38
5.4.8	Scatterometer (Optical Detector)	5-39

	5.4.9	Liquid Water Content Meter	5-41
	5.4.10	Drop Size Distribution Meter	5-43
	5.4.11	Optical Thermoelectric Dew Point Hygrometer	5-45
	5.4.12	Electric Dew Point Hygrometer	5-45
<b>5.5</b>		<b>Experiment Chambers and Aerosol Conditioning Subsystem</b>	<b>5-46</b>
	5.5.1	Static Diffusion Liquid (SDL) Chamber	5-48
	5.5.2	Static Diffusion Ice Chamber Assembly	5-56
	5.5.3	General Chamber	5-62
	5.5.4	Expansion Chamber Assembly	5-68
	5.5.5	Continuous Flow Diffusion Chamber Assembly	5-74
	5.5.6	Earth Simulation Chamber Assembly	5-80
	5.5.7	Nuclei Conditioning Assembly	5-85
<b>5.6</b>		<b>Console</b>	<b>5-89</b>
	5.6.1	Console Support Structure and Subassembly	5-90
	5.6.2	Power Control and Distribution	5-91
	5.6.3	Console Panels and Drawer Subassembly	5-96
	5.6.4	Instrumentation and Displays	5-99
<b>5.7</b>		<b>Optical and Imaging Devices</b>	<b>5-99</b>
	5.7.1	Cine Camera (35 mm)	5-101
	5.7.2	Still Camera (35 mm)	5-104
	5.7.3	Microscope Trinocular	5-104
	5.7.4	Video Camera Assembly	5-105
	5.7.5	Light Source	5-105
	5.7.6	Anemometer	5-106
	5.7.7	Stereo Microscope	5-107
	5.7.8	Infrared Microscope	5-108
	5.7.9	Support Equipment/Expendables	5-109
	5.7.10	Displays/Controls	5-110
<b>Section 6</b>		<b>LABORATORY SUPPORTIVE ANALYSES</b>	<b>6-1</b>
<b>6.1</b>		<b>Summary of Results</b>	<b>6-1</b>
	6.1.1	Aerosol Storage	6-2
	6.1.2	Chamber Operation	6-3
	6.1.3	Gravity Levels	6-4
	6.1.4	Aerosol Transport	6-4

6.1.5	Radiation Pressure and Heating	6-4
6.1.6	Air Ionization	6-4
6.1.7	Contamination	6-5
6.1.8	Air and Water Quantity	6-5
6.1.9	Cloud Chamber Thermal Design	6-5
6.2	Evaluation Details for the Phenomenological Factors	6-6
6.2.1	Diffusion (Brownian)	6-7
6.2.2	Phoretic Forces	6-8
6.2.3	Thermal (Brownian) Coagulation	6-13
6.2.4	Sedimentation	6-17
6.2.5	Convection	6-30
6.2.6	Acceleration Level (g-Level)	6-35
6.2.7	Aerosol Transport	6-37
6.2.8	Radiation Pressure	6-38
6.2.9	Radiation Heating	6-41
6.2.10	Air Ionization	6-43
6.3	Engineering Analysis	6-46
6.3.1	Contamination Assessment	6-47
6.3.2	Air and Water Quantity Requirements	6-53
6.3.3	Cloud Chamber Thermal Design	6-56
6.3.4	Cloud Chamber Optical Requirements	6-64

## Section 7

### TEST PHILOSOPHY AND PLAN AND SAFETY, RELIABILITY AND MAINTAINABILITY ANALYSES

		7-1
7.1	Test Philosophy and Test Plan	7-1
7.1.1	Test Philosophy	7-1
7.1.2	Test Plan	7-3
7.2	System Safety Plan	7-10
7.2.1	System Safety Program Plan	7-11
7.2.2	Hazard Review Checklist	7-13
7.2.3	Hazard Analysis	7-14
7.2.4	System Safety Plan for Phase B	7-20
7.3	Reliability Analysis	7-24
7.3.1	Purpose	7-24
7.3.2	Objective	7-24
7.3.3	Approach and Assumptions	7-24
7.3.4	Observations, Conclusions, and Recommendations	7-35
7.3.5	CPL Program Cost - Reliability Relationships	7-39
7.3.6	CPL Reliability Initial Goals and Subsystem Allocations	7-43

7.4	Maintainability Analysis	7-43
7.4.1	Approach	7-44
7.4.2	Cost Sensitivity Analyses	7-48
7.4.3	Recommendations	7-53

## Section 8 SUPPORTING RESEARCH AND TECHNOLOGY 8-1

8.1	Assessment and Recommendation	8-1
8.2	Technical Assessment	8-1
8.2.1	Priority	8-1
8.2.2	Cost	8-2
8.2.3	Schedule	8-3
8.3	Programmatic Assessment	8-3
8.3.1	Schedule Risk	8-5
8.3.2	Program Critical	8-6
8.4	Overall System Ranking	8-7
8.5	Supporting Research and Technology Categories	8-8
8.5.1	Research (R)	8-8
8.5.2	Advanced Technology (AT)	8-8
8.5.3	Advanced Development (AD)	8-8
8.5.4	Supporting Development (SD)	8-9
8.6	Supporting Research and Technology - Technology Areas	8-9
8.6.1	Acoustics/Acoustical	8-9
8.6.2	Fluid Dynamics	8-9
8.6.3	Electromechanical	8-9
8.6.4	Optics/Optical	8-9
8.6.5	Structural/Mechanical	8-10
8.6.6	Thermal	8-10
8.7	Supporting Research and Technology - Items	8-10

## Section 9 CLOUD LABORATORY SUPPORT OPERATIONS 9-1

9.1	CPL Operations	9-1
9.1.1	CPL Ground Operations	9-2
9.1.2	Functional Flow Diagrams	9-4
9.1.3	CPL Operational Schedule	9-4
9.2	Experiment Flight Support Analysis	9-13
9.2.1	PI Role in Experimental Support	9-14
9.2.2	Typical Flight Support Schedules	9-14
9.2.3	Simulator Requirements and Utilization	9-18
9.2.4	Initial Flight Support Operation Schedule	9-20

Section 10	DATA MANAGEMENT OPERATIONS PLAN	10-1
10.1	Mission Plans	10-1
10.2	Requirement Analysis	10-12
10.3	Processing Allocation and Flows	10-14
10.4	DMS Equipment Operation	10-17
10.4.1	Recording	10-19
10.4.2	Data Processing	10-21
10.4.3	Command	10-27
10.4.4	Controls	10-28
10.4.5	Data Transfer	10-31
10.4.6	Caution and Warning	10-32
10.4.7	Information Transfer	10-33
	Internal to the Laboratory	10-33
10.4.8	Data Dissemination	10-35
10.4.9	Data Formats	10-36

Section 11	CLOUD PHYSICS EXPERIMENT LABORATORY LABORATORY SCHEDULES	11-1
11.1	Summary	11-1
11.2	Schedule Guidelines	11-5
11.3	Project Schedule Factors	11-6
11.4	Laboratory Schedule	11-9
11.4.1	Supporting Research and Technology (SRT)	11-9
11.4.2	Interfacing Milestones	11-9
11.4.3	Cloud Physics Laboratory Milestones	11-9
11.4.4	Project Management	11-10
11.4.5	System Engineering and Integration	11-10
11.4.6	Cloud Physics Laboratory	11-10
11.4.7	Experiment Support Hardware	11-12
11.4.8	System Test	11-12
11.4.9	Ground Support Equipment	11-13
11.4.10	Facilities	11-13
11.4.11	Logistics	11-13
11.4.12	Ground Operations	11-14
11.4.13	Flight Operations for Cloud Physics Laboratory Launches	11-14
11.4.14	Principal Investigator Operations	11-14
11.5	Subsystem Schedules	11-15

REFERENCES	R-1
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Appendix A	WORK BREAKDOWN STRUCTURE AND DICTIONARY FOR ZERO GRAVITY CLOUD PHYSICS EXPERIMENT LABORATORY DEFINITION STUDY	A-1
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## VOLUME II

<b>Section 1</b>	<b>SUMMARY</b>	<b>1-1</b>
<b>Section 2</b>	<b>EXPERIMENT PROGRAM SUMMARY</b>	<b>2-1</b>
	2.1 Experiment Program	2-3
	2.2 Cloud Chamber Selection	2-11
	2.3 Research Related Equipment	2-17
	2.4 Mission Assessment	2-17
	2.5 Priority Mission Ranking	2-23
	2.6 Priority Mission Set – Total Experiment Program Requirements Comparison	2-25
<b>Appendix A</b>	<b>CONDENSATION NUCLEATION</b>	<b>A-1</b>
<b>Appendix B</b>	<b>ICE NUCLEATION</b>	<b>B-1</b>
<b>Appendix C</b>	<b>ICE MULTIPLICATION</b>	<b>C-1</b>
<b>Appendix D</b>	<b>CLASS 4 CHARGE SEPARATION (ELECTRIFICATION)</b>	<b>D-1</b>
<b>Appendix E</b>	<b>CLASS 5 ICE CRYSTAL GROWTH HABITS</b>	<b>E-1</b>
<b>Appendix F</b>	<b>CLASS 6 SCAVENGING</b>	<b>F-1</b>
<b>Appendix G</b>	<b>CLASS 7 RIMING AND AGGREGATION</b>	<b>G-1</b>
<b>Appendix H</b>	<b>CLASS 8 DROPLET-ICE CLOUD INTERACTIONS</b>	<b>H-1</b>
<b>Appendix I</b>	<b>CLASS 9 HOMOGENEOUS NUCLEATION (ICE)</b>	<b>I-1</b>
<b>Appendix J</b>	<b>CLASS 10 COLLISION-INDUCED FREEZING</b>	<b>J-1</b>
<b>Appendix K</b>	<b>CLASS 11 SATURATION VAPOR PRESSURE (SUPERCOOLED WATER)</b>	<b>K-1</b>
<b>Appendix L</b>	<b>CLASS 12 ADIABATIC CLOUD EXPANSION</b>	<b>L-1</b>
<b>Appendix M</b>	<b>CLASS 13 ICE NUCLEI MEMORY</b>	<b>M-1</b>
<b>Appendix N</b>	<b>CLASS 14 TERRESTRIAL EXPANSION CHAMBER EVALUATION</b>	<b>N-1</b>
<b>Appendix O</b>	<b>CLASS 15 CONDENSATION NUCLEI MEMORY</b>	<b>O-1</b>
<b>Appendix P</b>	<b>CLASS 16 NUCLEI MULTIPLICATION</b>	<b>P-1</b>

Appendix Q	CLASS 17 DROP COLLISION BREAKUP ( $< 0.5 \text{ nm}$ )	Q-1
Appendix R	CLASS 18 COALESCENCE EFFICIENCY ( $< 5.0 \text{ }\mu\text{m}$ )	R-1
Appendix S	CLASS 19 STATIC DIFFUSION CHAMBER EVALUATION	S-1
Appendix T	CLASS 20 UNVENTILATED DROPLET DIFFUSION COEFFICIENTS	T-1
Appendix U	CLASS 21 EARTH SIMULATION	U-1

# **FIGURES VOLUME 1**

<b>1</b>	<b>Space Shuttle Rendering</b>	<b>ix</b>
<b>2</b>	<b>Cloud Physics Application Area</b>	<b>xii</b>
<b>3</b>	<b>CPL Inboard View</b>	<b>xv</b>
<b>4</b>	<b>CPL Outboard View</b>	<b>xvi</b>
<b>5</b>	<b>Cloud Chambers</b>	<b>xix</b>
<b>2-1</b>	<b>Solicitation</b>	<b>2-2</b>
<b>3-1</b>	<b>Experiment Class Evaluations</b>	<b>3-3</b>
<b>3-2</b>	<b>Cloud Chambers</b>	<b>3-12</b>
<b>3-3</b>	<b>Effective Experiment Observation Times</b>	<b>3-18</b>
<b>3-4</b>	<b>Experiment Parameter Requirements</b>	<b>3-19</b>
<b>4-1</b>	<b>CPL Inboard View</b>	<b>4-7</b>
<b>4-2</b>	<b>CPL Outboard View</b>	<b>4-8</b>
<b>4-3</b>	<b>Cloud Physics Console Configuration</b>	<b>4-9</b>
<b>4-4</b>	<b>Static Diffusion Liquid Chamber Coefficient</b>	<b>4-12</b>
<b>4-5</b>	<b>Static Diffusion Ice Chamber</b>	<b>4-14</b>
<b>4-6</b>	<b>General Chamber Configuration</b>	<b>4-16</b>
<b>4-7</b>	<b>Expansion Chamber</b>	<b>4-18</b>
<b>4-8</b>	<b>Continuous Flow Diffusion Chamber</b>	<b>4-20</b>
<b>4-9</b>	<b>Earth Simulation Chamber Configuration</b>	<b>4-22</b>
<b>4-10</b>	<b>Typical Experiment Integration Procedure</b>	<b>4-40</b>
<b>5-1</b>	<b>Cloud Chamber Cooling Subassembly</b>	<b>5-4</b>

5-2	Flow, Humidity, and Pressure Control	5-6
5-3	Expendables Storage	5-7
5-4	Wire Probe Retractor Generator	5-14
5-5	Water Drop Impeller Generator	5-14
5-6	Vibrating Orifice Generator	5-16
5-7	Evaporator Condenser Generator	5-16
5-8	Spray Atomizer Generator	5-18
5-9	Powder Dispersion Generator	5-18
5-10	Particle Injector and Size Conditioner	5-19
5-11	Data Management Block Diagram	5-20
5-12	Spacelab DMS Equipment Diagram	5-22
5-13	Interface Adapter	5-25
5-14	Controllers	5-26
5-15	Optical Particle Counter	5-35
5-16	Condensation Nucleus Counter	5-37
5-17	Microporous Filter Subsystem	5-37
5-18	Piezoelectric Quartz Crystal Mass Monitor	5-38
5-19	Cascade Impactor	5-39
5-20	Electrical Aerosol Size Analyser	5-40
5-21	Scatterometer	5-42
5-22	Liquid Water Content Meter	5-43
5-23	Drop Size Distribution Meter	5-44
5-24	Optical Thermoelectric Dew Point Hygrometer	5-46
5-25	Electric Dew Point Hygrometer	5-47
5-26	Cloud Chambers	5-49
5-27	Static Diffusion Liquid Chamber	5-53

5-28	Static Diffusion Liquid Chamber	5-54
5-29	Static Diffusion Ice Chamber	5-57
5-30	Static Diffusion Ice Chamber	5-58
5-31	General Chamber	5-63
5-32	General Chamber	5-64
5-33	Expansion Chamber	5-69
5-34	Expansion Chamber	5-70
5-35	Continuous Flow Diffusion Chamber	5-77
5-36	Continuous Flow Diffusion Chamber	5-79
5-37	Earth Simulation Chamber	5-81
5-38	Earth Simulation Chamber	5-82
5-39	Nuclei Conditioning Chamber	5-85
5-40	Nuclei Conditioning Chamber	5-86
5-41	Total Cloud Physics Console/Cabinet Configuration	5-90
5-42	Structure Modification - 1.55-M and 0.572-M Cabinets	5-92
5-43	Power Distribution	5-93
5-44	28 VDC Regulated Circuits	5-94
5-45	110 VAC 3 Phase 400 Hz and 110 VAC 1 Phase 400 Hz Circuits	5-94
5-46	110 VAC 1 Phase 60 Hz Circuit	5-95
5-47	Volume Breakdown of 0.572-M Cabinet	5-97
5-48	Volume Breakdown of 1.55-M Cabinet	5-98
6-1	Velocity of Particles in Superimposed Thermal and Water-Vapor Pressure Gradients	6-9
6-2	Enhanced Velocity of Deposition in a Super-saturated Atmosphere	6-10
6-3	Thermo- and Diffusiophoretic Forces	6-11

6-4	Coagulation Rates for Monodisperse Particle Radius $a = 1.0 \mu\text{m}$	6-16
6-5	Coagulation Rates for Monodisperse Particle Radius $a = 0.005 \mu\text{m}$	6-16
6-6	Orbit Altitude Effect on Raindrop - X Axis Displacement	6-26
6-7	Orbit Altitude Effect on Raindrop - Y Axis Displacement	6-26
6-8	Orbit Altitude Effect on Raindrop - Z Axis Displacement	6-27
6-9	Particle Displacement in Various Package Orientations	6-28
6-10	Shuttle/Spacelab Acceleration Levels	6-36
6-11	Aerosol Transport Losses ( $r = 5 \times 10^{-7} \text{ cm}$ )	6-39
6-12	Aerosol Transport Losses ( $r = 10^{-5} \text{ cm}$ )	6-39
6-13	Humidification Chamber Weight	6-55
6-14	Effective Experiment Observation Times	6-56
6-15	Steady-State Thermoelectric Module Performance	6-63
6-16	Static Diffusion Ice Chamber Internal Wall Temperature Response	6-64
6-17	Phase Change Material Refreeze Time for 10 KG of $\text{C}_{15}\text{H}_{32}$	6-65
6-18	Standardized Chamber Optical Configuration	6-66
6-19	Standardized Chamber Optical Configuration	6-66
6-20	Coupling Optics Zoom Focus Capability	6-68
6-21	Illumination Module	6-69
6-22	Cross Lighting	6-70
7-1	Schedule Analysis Model Plan	7-7
7-2	Development Program Schedule Analysis Guide	7-8
7-3	Requirement/Model Matrix	7-9

7-4	Estimator Used to Relate System Acquisition Cost Trends to Program Equivalent Reliability Levels	7-27
7-5	Indicated Z (Reliability Level) for Minimum Variable CPL Program Costs vs Q (Unreliability Estimate)	7-29
7-6	CPL-Only Costs per Mission Estimates for Costs Variable with CPL Reliability Level	7-30
7-7	CPL Mission and S/L Plus Shuttle Mission Costs by Equivalent Reliability Level	7-31
7-8	CPL Total Mission Cost Estimates by Equivalent Reliability Level	7-33
7-9	CPL Total Mission Cost Estimates of Costs Variable by CPL Reliability Level for a Range of CPL Q Estimates and S/L and Shuttle Allocated Costs	7-34
7-10	CPL Cost as a Function of Reliability Level	7-38
7-11	Total M&R Costs for 42 Flights as a Function of Field Site M&R Manhours per Flight	7-49
7-12	M&R as a Percent of Total Operations Cost	7-52
7-13	Index of Total Operations Cost as a Function of Field Site M&R Manhours per Flight	7-53
8-1	Schedule Relationship Cloud Physics Laboratory Development and SRT	8-4
9-1	CPL Ground Operations Scenario	9-3
9-2	CPL Ground Operations Scenario	9-5
9-3	Postlanding Recovery Flow	9-7
9-4	Maintenance and Refurbishment Flow	9-8
9-5	Integration Checkout Flow	9-9
9-6	Prelaunch and Launch Checkout Flow	9-10
9-7	5.0 Mission Operations	9-11
9-8	Cloud Physics Laboratory Ground and Flight Operations	9-12
9-9	Typical Experiment Flight Support (11 Month)	9-15

9-10	Typical Experiment Flight Support (9 months)	9-17
9-11	CPL Flight Support Operations (Flights 3 to 7)	9-19
9-12	Flight Support Operations CPL Flights 1 and 2	9-21
10-1	Mission Plan 1 - Recover Data	10-3
10-2	Mission Plan 2 - DMS Inspection and Checkout	10-4
10-3	Mission Plan 3 - DMS Maintenance and Repair	10-5
10-4	Mission Plan 4 - Configure CPL (DMS) for Next Mission	10-6
10-5	Mission Plan 5 - DMS Functional Verification and Test	10-7
10-6	Mission Plan 6 - Integrate Lab, CPL (DMS)	10-8
10-7	Mission Plan 7 - CPL Power-Up, Interface Verification	10-9
10-8	Mission Plan 8 - CPL Experiment Operation	10-10
10-9	On-Orbit Data Processing Summary	10-12
10-10	CPL Function Allocation	10-14
10-11	Primary Experiment/Data Flow	10-15
10-12	Internal Processing Flows	10-16
10-13	Data Management Block Diagram	10-18
10-14	Aerosol Generation Timeline	10-22
10-15	Aerosol Generation Flow	10-23
10-16	Flow, Humidity, and Pressure Control	10-26
10-17	Regional Data Flow	10-37
10-18	Prime Frame Format	10-39
10-19	Graphic Display Formats	10-41
11-1	Cloud Physics Experiment Laboratory Project Schedule	11-2



11-2	Cloud Physics Experiment Laboratory Master Program Chart	11-3
11-3	Cloud Physics Experiment Laboratory Subsystems Schedule	11-4

## VOLUME II

1	Space Shuttle Rendering	ix
2	Cloud Physics Requirements	xii
3	CPL Inboard View	xvi
4	CPL Outboard View	xvii
5	Cloud Chambers	xix
2-1	Experiment Class Evaluations	2-3
2-2	Cloud Chambers	2-13
2-3	Effective Experiment Observation Times	2-19
2-4	Experiment Parameter Requirements	2-20
A-1	CFD Layout	A-13
A-2	Activity Timeline (One Day) Experimental Class 1	A-16
A-3	Continuous-Flow Diffusion Chamber	A-34
A-4	Continuous-Flow Diffusion Chamber Air View	A-34
A-5	Activity Timeline (One Day) Experiment Class 1 Condensation Nuclei	A-39
A-6	Activity Timeline (One Day) Experiment Class 1 Condensation Nuclei	A-50
B-1	Sketch of University of Denver Static Thermal Diffusion Chamber	B-5
B-2	Activity Timeline (One Day) Experiment Class 2	B-15
B-3	Activity Timeline (One Day) Experiment Class 2	B-24

C-1	Activity Timeline (One Day) Experiment Class 3	C-9
D-1	Activity Timeline (One Day) Experiment Class 4	D-7
E-1	Activity Timeline (One Day) Experiment Class 5	E-8
E-2	Activity Timeline (One Day) Experiment Class 5	E-16
F-1	Activity Timeline (One Day) Experiment Class 6	F-14
G-1	Activity Timeline (One Day) Experiment Class 7	G-8
G-2	Activity Timeline (One Day) Experiment Class 7	G-15
H-1	Activity Timeline (One Day) Experiment Class 8	H-8
I-1	Activity Timeline (One Day) Experiment Class 9	I-8
I-2	Activity Timeline (One Day) Experiment Class 9	I-17
I-3	Activity Timeline (One Day) Experiment Class 9	I-34
J-1	Activity Timeline (One Day) Experiment Class 10	J-6
K-1	Activity Timeline (One Day) Experiment Class 11	K-8
K-2	Activity Timeline (One Day) Experiment Class 11	K-18
L-1	Activity Timeline (One Day) Experiment Class 12	L-14
M-1	Activity Timeline (One Day) Experiment Class 13	M-10
M-2	Process of Adiabatic Expansion Chamber for Memory Study	M-11
M-3	Activity Timeline (One Day) Experiment Class 13	M-24
N-1	Activity Timeline (One Day) Experiment Class 14	N-6
O-1	Activity Timeline (One Day) Experiment Class 15	O-9
O-2	Activity Timeline (One Day) Experiment Class 15	O-22
P-1	Activity Timeline (One Day) Experiment Class 16	P-14
P-2	Purge System Schematic	P-22
P-3	Humidifier	P-22
P-4	Activity Timeline (One Day) Experiment Class 16	P-27
Q-1	Activity Timeline (One Day) Experiment Class 17	Q-11

Q-2	General Purpose Chamber	Q-12
Q-3	Activity Timeline (One Day) Experiment Class 17	Q-20
R-1	Activity Timeline (One Day) Experiment Class 18	R-14
R-2	Activity Timeline (One Day) Experiment Class 18	R-21
S-1	Activity Timeline (One Day) Experiment Class 19	S-9
T-1	Activity Timeline (One Day) Experiment Class 20	T-11
T-2	Kohler Curve	T-22
T-3	The Curvature Effect Evolution	T-22
T-4	Activity Timeline (One Day) Experiment Class 20	T-34
U-1	Schematic of Spherical Convection Chamber	U-9
U-2	Block Schematic of Terrestrial Spherical Convection Experiment Showing Control Equipment Which is Connected to the Chamber Through Slip Rings in the Turntable Base	U-10
U-3	Block Schematic of Space Laboratory Version of the Spherical Convection Experiment	U-13
U-4	Activity Timeline (One Day) Experiment Class 21	U-18
U-5	Activity Timeline (One Day) Experiment Class 21	U-19

## **Section 1**

### **SUMMARY**

**This study summarizes work accomplished from January 1974 to October 1974 for the Zero-Gravity Atmospheric Cloud Physics Laboratory. This project involves the definition and development of an atmospheric cloud physics laboratory and the selection and delineation of a set of candidate experiments that require the unique environment of zero gravity or near zero gravity.**

**General objectives of the Zero-Gravity Atmospheric Cloud Physics Laboratory program are to significantly increase the level of knowledge in atmospheric cloud physics research by placing at the disposal of the terrestrial-bound atmospheric cloud physicist a laboratory that can:**

- A. Complement and/or supplement the cloud physics research performed in terrestrial laboratories.**
- B. Incorporate design features which would ensure facility usage over the largest feasible range of research and beneficial application experimentation.**
- C. Provide a unique laboratory for cloud physics researchers, eliminating the use of mechanical, aerodynamic, electrical, and other techniques which tend to mask data results.**

**Scientific objectives of the Zero-Gravity Atmospheric Cloud Physics Laboratory project are: to advance the state of the knowledge in atmospheric cloud microphysics, to provide an unique laboratory for cloud physics researchers, and to develop techniques in weather control and modification.**

**Cloud physics research under zero- or low-gravity conditions offers an opportunity to answer many problems that cannot be solved in earth-based laboratories. By taking advantage of zero gravity to define many of the processes in clouds that are not yet fully understood, man could influence weather by changing, for example, drop distributions and nuclei**

concentrations, or by adding pollutant compositions. Under zero gravity, an experimenter can suspend a drop in a chamber and observe its nature for the actual time required for various processes and forces to take effect. The droplet can be frozen and thawed out, and another drop can be propelled into it. Observations can be made of the migration and collection of particulate matter that may be near or around the drop. These characteristics cannot be investigated on earth because of gravity and, in some instances, because of effects of measures taken to offset gravity. Thus, numerous experiments that cannot be done on earth can be performed in this unique environment. The laboratory will be made available to the entire cloud physics community so that a wide variety of important experiments can be performed. Participation of the scientific community was encouraged, supporting research was done at universities, and many valuable suggestions by scientists in industry, Government, and universities were incorporated in the concept.

The results of the study are presented in two Volumes:

- Volume I - presents the results of the total study effort
- Volume II - presents the detailed approaches identified for each class of the experiment program

The primary contents of Volume I are described in the following. In Section 2 of that volume, the three phases of the feasibility study (reported in NASA CR 128998, NASA CR 129002, and NASA CR 129013) are reviewed. The first phase of the study accomplished the following:

- A. Developed scientific community support interface.
- B. Solicited experiment definitions.
- C. Selected high-priority experiments.
- D. Determined program feasibility.
- E. Identified a concept for multiple experiment cloud physics laboratory including subsystems and components.

The second phase of the study accomplished the following:

- A. Experiments suggested by the scientific community were defined and classified.

- B. Twenty classes of such experiments that require zero or low gravity were identified.
- C. Laboratory requirements were determined, based on the experiment class definitions.
- D. A multiple experiment laboratory concept was established to accommodate nearly all the experiments.

The final phase of the study accomplished the following:

- A. Experiment program revisions based on scientific community and Senior Scientific Board evaluation and critique.
- B. Establishment of three experiment mission timelines.
- C. Formulation of the laboratory concept and major subsystems based on experiment class requirements.
- D. Assessment of project technical risk including identification of required supporting research and technology (SRT) items.
- E. Formulation of key programmatic aspects of the project.

Section 3 presents the summary of the presently defined experiment program. The 20 experiment classes for cloud microphysics and the macroscale experiment class for earth simulation are identified with their objectives. The primary and alternate cloud chambers for such experiment class are identified. A summary of the range of experiment timelines is also presented. The results of the analysis to establish the primary and secondary variables are presented by experiment class. A priority mission set evaluation and the total experiment program mission assessment are also presented.

Section 4 describes the rationale, guidelines, constraints, and design features used to formulate the laboratory concept. The laboratory description includes total laboratory characteristics and the subsystems, including their use and location. Interfaces with the Spacelab, CPL growth, and scientific community participation are included.

Section 5 describes the individual laboratory subsystems, their assemblies and components and use by experiment class.

Section 6 contains summaries of the major analyses and tradeoffs performed in support of the study. These analysis include the evaluation of phenomenological factors influencing experiment program conduct. Engineering analyses include the evaluation of contamination/cleanliness, expendable quantities, thermal control, and optics design features.

Section 7 is comprised of four separate sections describing the test philosophy and test plan, preliminary laboratory safety plan, reliability analysis, and maintainability analysis.

Section 8 describes the Supporting Research and Technology (SRT) items identified for the CPL. The SRT assessment methodology and priority ranking are defined.

Section 9 defines the anticipated CPL operations. The project aspects of operations scheduling, laboratory maintenance, refurbishment and reconfiguration, and principal investigator participation are presented.

Section 10 includes the data management and processing plan. The CPL data processing requirements and their use in establishing data flow to both the principal investigators and to the CPL operations contractor is defined. Identification of Shuttle-Spacelab support and interfaces are included in the plan.

Section 11 defines the major milestones for the CPL project, the laboratory, project schedule and the supporting subsystem schedules. The schedule driving factors are presented for all aspects of the project.

The Appendix A contains the Work Breakdown Structure (WBS) and Dictionary for the project. This WBS reflects the status of the laboratory and the project as a result of the efforts conducted in the study.

The primary contents of Volume 2 are described in the following.

Section 2 presents the summary of the presently defined experiment program (as provided in Volume 1 section 3).

Appendixes A through U present the detailed experiment class descriptions for Experiment Classes 1 through 21. The detailed descriptions contain the identification of experiment objectives, justification, applications, approach, limitations, and timelines.



## Section 2

### EXPERIMENT PROGRAM SUMMARY

The experiment viability and scientific usefulness of the zero-gravity CPL program relies heavily on the interest and participation of active cloud physics scientists. This participation was reflected by the involvement of the individuals identified in Table 2-1. These individuals from the scientific community contributed extensively to the reevaluation and refinement of the experiment program as was defined in the NASA Contract Report CR129013 of September 1973. Their input included the delineation of scientific and project pertinent data.

The scientific data included experiment class objective, justification, applications, limitation, and approach as well as the value of the orbital near-zero-gravity experiment opportunity and quantification of pertinent experiment characteristics, parameters, and requirements. Project data included delineation of the groups and parameters within the experiment class, a typical mission timeline, and consumable usage. The format of such experiment descriptions is shown in Figure 2-1 along with significant results of such descriptions.

In general, two independent scientific evaluations were obtained for each experiment class. These inputs were used to establish the CPL Experiment Program and to extract experiment requirements for the CPL. The following subsections identify the Experiment Program and the results of major efforts performed using the Experiment Class descriptions as basic data. Volume II of this report contains an edited version of the scientific community contributions to the Experiment Class Descriptions.

In addition to these major contributors, a survey of project interest and participation was conducted of the general scientific community areas (cloud

Table 2-1  
EXPERIMENT PROGRAM CONSULTANTS

---

Prof. C. Anderson, U. of Wisconsin
Mr. J. Anderson, U. of Nevada
Dr. D. Blanchard, SUNY
Dr. J. Carstens, U. of Missouri
Dr. R. de Pena, Penn State University
Prof. N. Fukuta, U. of Denver
Prof. N. Gokhale, SUNY
Prof. J. Hallett, U. of Nevada
Dr. J. Hart, MIT
Prof. T. Hoffer, U. of Nevada
Dr. K.O.L.F. Jayaweera, U. of Alaska
Prof. J. Kassner, U. of Missouri
Dr. U. Katz, CALSPAN
Dr. W. Kocmond, CALSPAN
Dr. G. Langer, NCAR
Dr. J. Lodge, NCAR
Dr. J. Podzimek, U. of Missouri
Dr. J. Spengler, Harvard
Prof. P. Squires, U. of Nevada
Dr. T. Takahashi, U. of Hawaii
Prof. J. Telford, U. of Nevada
Dr. D. White, U. of Missouri

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modeling, field experimentation, and laboratory research). A solicitation was sent to 86 scientists. Of the 72 replies received, 63 indicated active interest, 9 of which promised and/or supplied experiment suggestions and descriptions. Responses indicating either a change of scientific area or of lack of interest in the project were received from nine individuals. The survey did prompt requests for additional data and reports and, in general, indicated a high degree of interest and desire to participate in the CPL Project.

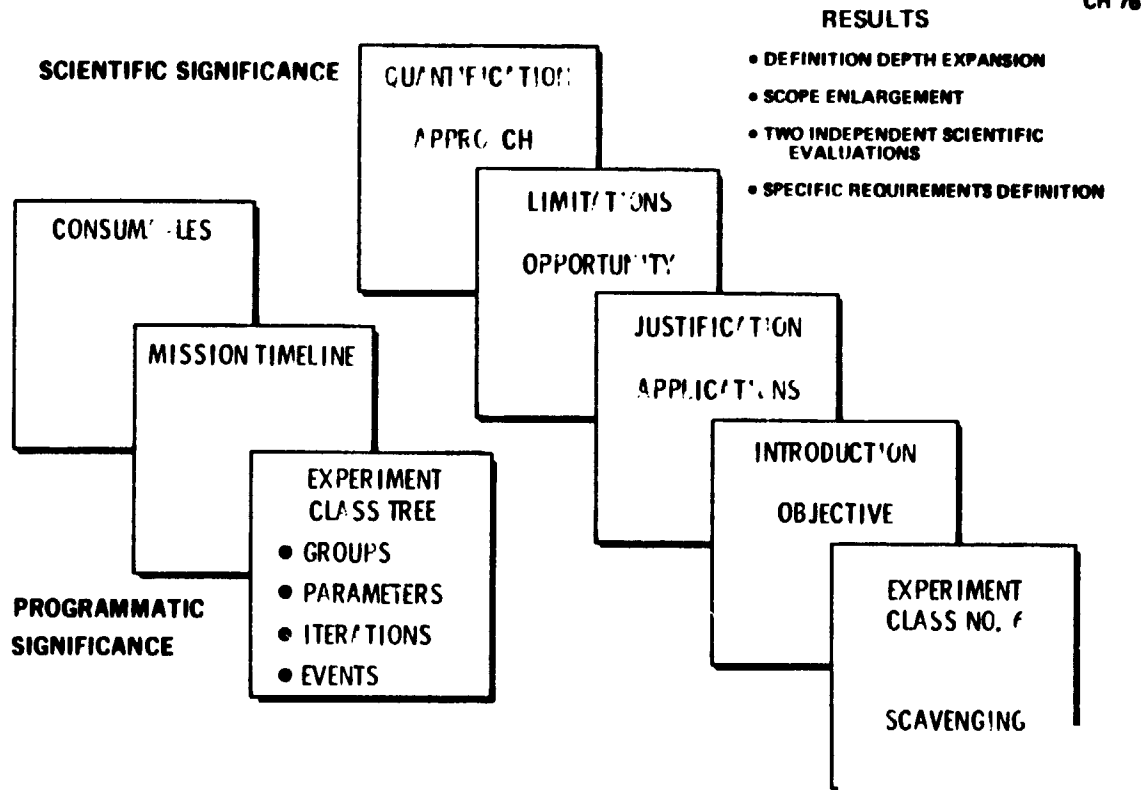


Figure 2-1. Experiment Class Evaluations

## 2.1 EXPERIMENT PROGRAM

The Cloud Physics Laboratory concept, presented later in this report, was developed to accommodate and satisfy the requirements of the experimentation selected and defined by the scientific community participants. The Experiment Program, as presently formulated, is summarized by Experiment Class Title and Objectives in Table 2-2. This table shows the breadth of experimentation planned to date.

These experiments relate to various phases of precipitation which mainly involve growth by ice and liquid particle collision and adherence through riming, clustering, and coalescence. Collision processes require relative velocities between particles which in turn requires differences in sizes or geometric shapes. These differences are generally due to varying parameters such as condensation nuclei characteristics and humidity distribution.

Most practical weather modification techniques are concerned with the production of a few large ice or water particles in a cloud of many smaller

Table 2-2 (Page 1 of 4)  
EXPERIMENT CLASSES AND OBJECTIVES

Experiment ID No.	Experiment Class Title	Objective
1	Condensation Nucleation	Determine the nucleation efficiencies and early growth properties of soluble, insoluble, and hydrophobic nuclei. This class of experiments encompasses a large range of nuclei types, size, distributions, and relative humidities.
2	Ice Nucleation	Determine the relative importance of contact, internal, and sublimation nucleation of ice. Absolute nucleation efficiencies will also be studied as a function of nuclei types and sizes.
3	Ice Multiplication	Determine the conditions under which ice fragments are generated during atmospheric precipitation processes and the extent to which they are generated.
4	Charge Separation (Electrification)	Determine quantitative values for charge transfer occurring during several important atmospheric processes.
5	Ice Crystal Growth Habit	Determine the temperature, pressure, and relative humidity conditions which dictate ice crystal geometry and growth rate under pure diffusion (nonconvective) conditions.
6	Scavenging	Determine the relative and quantitative importance of thermal (thermophoresis), diffusional (diffusiophoresis), and Brownian forces in the collection of submicrometer aerosol particles by cloud droplets and ice crystals.
7	Riming and Aggregation	Determine interaction between a supercooled water droplet and an ice surface during events associated with riming and graupel formation.

Table 2-2 (Page 2 of 4)  
EXPERIMENT CLASSES AND OBJECTIVES

Experiment ID No.	Experiment Class Title	Objective
8	Droplet-Ice Cloud Interactions	Determine the modes and extent of the interactions of ice crystals and supercooled water droplets, including the propagation of the ice phase through a supercooled droplet cloud and the diffusional growth of ice crystals within a cloud of supercooled droplets under varying conditions of temperature, pressure, and droplet/crystal concentrations.
9	Homogeneous Nucleation (Ice)	Determine the homogeneous freezing distribution of droplets as a function of time, degree of supercooling, and droplet diameter under conditions of no physical supports.
10	Collision-Induced Freezing	Determine the conditions and frequency of droplet freezing due to collisions of supercooled droplets as a function of droplet size, impact energy, and various ambient conditions of temperature, pressure, and relative humidity. Effects of electric and sonic fields will also be investigated.
11	Supercooled-Water Saturation Vapor Pressure	Determine the saturation vapor pressure of supercooled water.
12	Adiabatic Cloud Expansion Simulation	Duplicate in time and conditions the early portion of the life cycle of a parcel of air involved in an atmospheric precipitation process.
13	Ice Nuclei Memory	Determine the effect of an ice nuclei's history on its ability to initiate (nucleate) the ice phase.

Table 2-2 (Page 3 of 4)  
EXPERIMENT CLASSES AND OBJECTIVES

Experiment ID No.	Experiment Class Title	Objective
14	Terrestrial Expansion Chamber Evaluation	Measure condensation and ice nuclei activation efficiencies under operating conditions similar to those utilized in terrestrial laboratories, but without gravity-induced convection.
15	Condensation Nuclei Memory	Determine the effect of a condensation nuclei's history on its ability to initiate (nucleate) the liquid phase.
16	Nuclei Multiplication	Determine the processes and extent of nuclei material breakup.
17	Droplet Collision Breakup	Determine the energy requirements of large droplet-droplet collision-induced breakup as a function of fluid properties, droplet diameters, and external field conditions (sound and electrical).
18	Coalescence Efficiencies	Determine the coalescence efficiencies of small ( $<50 \mu\text{m}$ ) cloud droplets under varying impact conditions with specific attention toward what happens at the droplet-droplet interface just before and during collision.
19	Static Diffusion Chamber Evaluation	Determine the absolute nucleation efficiencies of standardized nuclei sources utilizing zero fallout conditions.
20	Unventilated Droplet Diffusion Coefficient	Determine the undisturbed diffusion (nonconvective) heat and mass transfer coefficients for growing and evaporating droplets (diameter greater than $10 \mu\text{m}$ ) under various conditions of temperature, pressure, and relative humidity and for various droplet diameters. This class of experiments will include the effects of various atmospheric contaminants on these coefficients.

Table 2-2 (Page 4 of 4)  
EXPERIMENT CLASSES AND OBJECTIVES

Experiment ID No.	Experiment Class Title	Objective
21	Earth Simulation	Determine the circulation and instability dynamics of the atmosphere and of the oceans by scaled and simulated experiments.

particles which thus initiates the collision process. These large particles are produced by their enhanced diffusion growth resulting from the lowering of their saturation vapor pressure below that of the ambient vapor pressure. This supersaturation condition can be produced in warm clouds by the addition of giant salt particles which results in a low vapor pressure salt solution. Dry ice (solid  $\text{CO}_2$ ) and various ice nucleating agents (e.g., AgI) are used in supercooled droplet clouds to produce a few frozen droplets with a corresponding lower saturation vapor pressure. In both of these cases, the modified particles grow at an accelerated rate, depending on temperature, humidity and relative numbers of modified to unmodified droplets.

There are four important areas of processes in clouds which must be better understood before productive deliberate modeling and weather prediction and modification can occur. These are nucleation, growth, scavenging, and electrical charge separation and the various classes of experiments can be loosely grouped into these four areas.

- A. Nucleation - Nucleation in cloud physics refers to the process of initiating the liquid or ice phase of water. Water vapor (free of ions and particulates) will not form a liquid phase unless a high supersaturation exists and the liquid will not freeze until it is cooled to below  $-35^\circ\text{C}$ . These two conditions for homogeneous nucleation do not exist under normal atmospheric conditions, but they are of theoretical interest as a foundation for the understanding of the general heterogeneous nucleation processes.

The normal atmosphere contains particles below 1.0- $\mu$ m diameter that remain suspended due to their negligible fall velocities. The number of these particles between 0.01 and 1.0  $\mu$ m available to serve as condensation nuclei is sufficient to limit the normal atmospheric supersaturation to well below 1 percent (relative humidity of 101.00 percent).

Particles larger than 1  $\mu$ m are generally referred to as "giant nuclei" and are limited in number due to gravitational fallout and because they are the first nuclei to become active in water droplet formation. Giant nuclei are provided artificially for warm cloud modification.

Ice nuclei are much more limited in numbers than condensation nuclei because of their special physical requirements. Cloud seeding often uses the supercooled condition that results from this shortage of ice nuclei.

Laboratory investigations have shown that once certain particles have acted as nucleating sites for water or ice, their activation characteristics are changed. This phenomenon is known as an ice and condensation nuclei memory effect.

Nucleation processes are involved in all forms of weather. At the present time, most weather modification involves the manipulation of nuclei (cloud seeding) within a given weather system. Current research is aimed at determining the role of the various atmospheric nuclei parameters (number, composition, effectiveness, and sources, including pollutants). Further understanding of the role of nuclei will aid in modification efforts such as: the increase of snow and rain for city and agricultural use; the decrease of destruction by hurricanes and hail; and the dissipation of airport and highway fog and smog. Basic to such modifications is knowledge of the nuclei to use, the appropriate number to introduce, the proper injection region in the weather system, and the optimum injection time during the development cycle.

- B. Growth - Once nucleation has been initiated, liquid or ice grows by condensation (vapor diffusion) until the particle reaches a few tens of micrometers in size. The quantitative values of the various thermal



and vapor accommodation coefficients are very important to this initial diffusional growth phase. Above 20- $\mu$ m diameter, field observations and theoretical computations indicate that other growth processes in addition to diffusion must be involved in order to explain the growth of particles to millimeter size in reasonable times, where they are able to fall from clouds as precipitation.

Included here are processes such as collision, coalescence (merging of two droplets), aggregation, and riming. These processes require a coexistence of particles (liquid or ice) with a range of sizes. Studies of the growth rates during various phases of growth are an important area of laboratory research and include: diffusional growth under normal atmospheric supersaturations (relative humidities below 101.0 percent), and freezing with possible breakup (splintering) as related to growth processes.

The study of growth processes is important in the "when and where" questions of weather modification while splintering affects the quantities of nucleating materials required.

- C. Scavenging — Droplets and ice crystals greater than a few micrometers in diameter collect (scavenge) gases, radioactive particles, and other atmospheric constituents. There is a continuing process of "washing-out" or cleansing of the atmosphere.

Particles below a few micrometers in diameter are collected by several processes, including those associated with Brownian motion, temperature gradients during evaporation (thermophoresis), vapor transport during condensation (diffusiophoresis), gravity-induced collisions (inertial), and electrical forces on charged particles. Normal fallout removes particles greater than 20  $\mu$ m in diameter. Scavenging is important in connection with ice nucleation efficiencies relative to weather modifications techniques and wash-out efficiencies as related to air pollution problems.

- D. Electrical Charge Separation — Cloud physicists are concerned with the processes of obtaining charge separation within natural clouds. Lab-

oratory investigations are concerned with charge transfer processes that occur during collision of ice with liquid or ice. Better understanding of electrical processes has potential in such areas as the reduction of forest fires and property damage due to lightning, and the assessment of the role of electrical phenomena in growth and scavenging processes.

It should be noted that the CPL concept is intended to accommodate not only the experimentation already identified but also be capable of flexibility to accommodate expanded experiment requirements not yet identified. The Experiment Program is, therefore, not yet complete or finalized. Additional project efforts include opportunities for inclusion of experiment suggestion.

## 2.2 CLOUD CHAMBER SELECTION

The Experiment Classes listed in Table 2-2 provided a working basis for developing experiment requirements. Of prime interest was the designation of the cloud chambers to be used in the Experiment Program.

The individuals identified in Table 2-1 designated their selection of both primary and alternate cloud chambers for each Experiment Class. The primary chamber designation indicated that the major objective of the Experiment Class could be evaluated best in that chamber. The alternate chamber designation indicated that information complementary to the major objective of the Experiment Class could be obtained in that chamber and/or that specific aspects of the Experiment Class necessitated its use. It should be noted that all contributors did not concur on primary and alternate cloud chamber designations and also that more than one alternate cloud chamber exists for many Experiment Classes.

The evaluation of the primary and alternate cloud chambers resulted in the selections shown in Table 2-3. The primary chamber designations are in concert with those identified in the Feasibility Study (NAS 8-27861). The alternate cloud chamber designations reflect reduction to a single alternate cloud chamber for each Experiment Class. The designations, in general, are based on the consensus of the scientific community recommendations. These recommendations were, however, impacted by consideration of

Table 2-3  
PRIMARY AND ALTERNATE CHAMBER SELECTION

Class No.*	Class Title	Primary Chamber	Alternate Chamber
1	Condensation Nucleation	CFD	E
2	Ice Nucleation	SDI	E
3	Ice Multiplication	SDI	E
4	Charge Separation	SDI	G
5	Ice Crystal Growth Habits	SDI	E
6	Scavenging	SDI	G
7	Riming and Aggregation	SDI	G
8	Droplet-Ice Cloud Interactions	SDI	E
9	Homogeneous Nucleation	SDI	E
10	Collision-Induced Freezing	SDI	G
11	Saturation Vapor Pressure	SDI	E
12	Adiabatic Cloud Expansion	E	-
13	Ice Nuclei Memory	E	SDI
14	Terrestrial Expansion Chamber Evaluation	E	-
15	Condensation Nuclei Memory	E	SDL
16	Nuclei Multiplication	G	E
17	Drop Collision Breakup	G	SDI
18	Coalescence Efficiencies	G	SDI
19	Static Diffusion Chamber Evaluation	SDL	-
20	Unventilated Droplet Diffusion Coefficients	SDL	E
21	Earth Simulation	ES	-

\*Ordered for data reduction purposes, not by priority

experiment operations, space environment considerations and cloud chamber specification requirements. These considerations generally resulted in alternate cloud chamber selection most compatible with the capability of the cloud chamber (designated based on its primary usage).

The cloud chambers to be used in CPL experimentation are shown in Figure 2-2. The pertinent geometric features, the significant operational features, the use, and the primary requirements are also identified in this figure. The requirements specified are compatible with the Experiment Class usage as primary and alternate cloud chambers.

Each of the six chambers that have been identified for use in zero-gravity CPL possess unique characteristics that enhance the investigation of certain

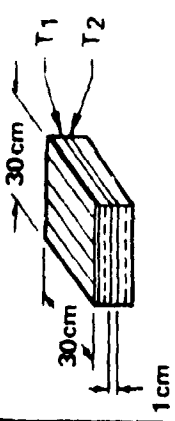
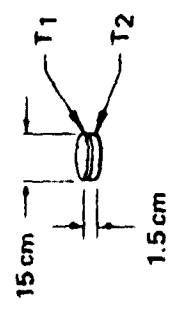
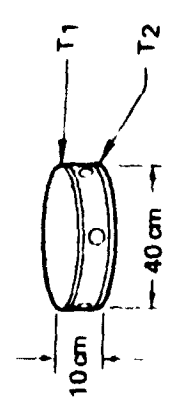
	 <p>CONTINUOUS-FLOW DIFFUSION (CFD)</p> <p><math>T_2 &lt; T_1</math> LIQUID SURFACES <math>0.01 \mu\text{m} &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 10 \mu\text{m}</math> OUTPUT - SIZE DISTRIBUTION CONDENSATION NUCLEATION STUDIES</p>	 <p>STATIC DIFFUSION, LIQUID (SDL)</p> <p><math>T_2 &lt; T_1</math> LIQUID SURFACES <math>0.01 \mu\text{m} &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 10 \mu\text{m}</math> OUTPUT - NUMBERS ONLY CONDENSATION NUCLEATION STUDIES</p>	 <p>STATIC DIFFUSION, ICE (SDI)</p> <p><math>T_2 &lt; T_1</math> ICE SURFACES <math>1 \mu\text{m} &lt; \text{PARTICLE}</math> DIAMETER <math>&lt; 1 \text{cm}</math> OUTPUT - SIZE, SHAPE ICE CRYSTAL STUDIES</p>	
	PRESSURE RANGE TOLERANCE	760 TO 700 mm ABS $\pm 5 \text{ mm}$ REL $\pm 1 \text{ mm}$	760 TO 140 mm ABS $\pm 10 \text{ mm}$ REL $\pm 1 \text{ mm}$	760 TO 140 mm ABS $\pm 10 \text{ mm}$ REL $\pm 1 \text{ mm}$
	RELATIVE HUMIDITY TOLERANCE	100% TO 103% ABS $\pm 0.02\%$ REL $\pm 0.01\%$	100% TO 103% ABS $\pm 1\%$ REL $\pm 0.5\%$	80% TO 120% ABS $\pm 1\%$ REL $\pm 0.5\%$
	TEMPERATURE TOLERANCE	$0^\circ\text{C}$ TO $+35^\circ\text{C}$ ABS $\pm 0.1^\circ\text{C}$ REL $\pm 0.02^\circ\text{C}$	$0^\circ\text{C}$ TO $+35^\circ\text{C}$ ABS $\pm 0.1^\circ\text{C}$ REL $\pm 0.02^\circ\text{C}$	$-60^\circ\text{C}$ TO $+35^\circ\text{C}$ ABS $\pm 1^\circ\text{C}$ REL $\pm 0.1^\circ\text{C}$
	$\Delta T (T_1 - T_2)$	0 TO $10^\circ\text{C}$	0 TO $10^\circ\text{C}$	0 TO $60^\circ\text{C}$
FLOW RATE	0.25 SCFM			

Figure 2-2. Cloud Chambers (Sheet 1 of 2)

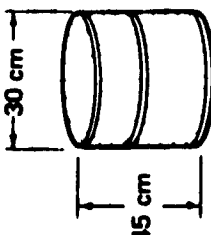
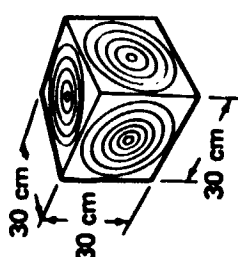
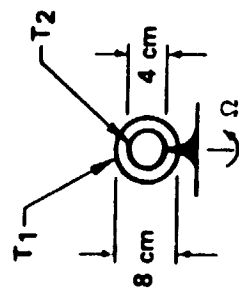
	<p>EXPANSION (E)</p> <p>HYDROPHOBIC SURFACES</p> <p>COOLED WALLS</p> <p>0.01 <math>\mu\text{m}</math> &lt; PARTICLE DIAM.</p> <p>ETER &lt; 100 <math>\mu\text{m}</math></p> <p>ADIABATIC EXPANSION</p> <p>OUTPUT - NUMBERS, MEAN SIZE</p>		<p>GENERAL (G)</p> <p>ELECTRIC FIELDS</p> <p>DROPLETS &gt; 100 <math>\mu\text{m}</math></p> <p>LARGE-PARTICLE INTERACTIONS</p>		<p>EARTH SIMULATION (ES)</p> <p>DIFFERENTIALLY HEATED</p> <p>ROTATING SPHERICAL</p> <p>ANNULUS OF FLUID</p> <p>ROTATION RATE - 6 RAD/SEC</p> <p>PLANETARY AND SOLAR</p> <p>CONVECTION STUDIES</p>
PRESSURE RANGE TOLERANCE	760 TO 140 mm ABS $\pm$ 2 mm REL $\pm$ 0.1 mm	760 TO 140 mm ABS $\pm$ 10 mm REL $\pm$ 1 mm	AMBIENT N/A		
RELATIVE HUMIDITY TOLERANCE	1 TO 100% ABS $\pm$ 1% REL $\pm$ 0.5%	1 TO 95% ABS $\pm$ 1% REL $\pm$ 0.5%	N/A - -		
TEMPERATURE TOLERANCE	-40°C TO +35°C ABS $\pm$ 0.5°C REL $\pm$ 0.05°C	-10°C TO +35°C ABS $\pm$ 1°C REL $\pm$ 0.1°C	+15°C TO +35°C ABS $\pm$ 0.1°C REL $\pm$ 0.03°C		
$\Delta T$ (T1 - T2)	-	-	0 TO 7°C		

Figure 2-2. Cloud Chambers (Sheet 2 of 2)

aspects of several cloud physics phenomena. On the other hand, a given phenomenological process can be viewed and studied from several approaches utilizing several chambers. These factors were included in the establishment of primary and secondary chamber assignments. The following paragraphs are presented to provide the salient features of each chamber.

**Continuous Flow Diffusion Chamber (CFD)** - This chamber provides a supersaturated environment for temperatures warmer than the freezing point of water. This chamber provides a rapid throughput of information and provides information on sizes of nuclei and droplets above  $0.3 \mu\text{m}$  in diameter. The chamber characteristics are ideally suited to the study of condensation nucleation and initial diffusional growth of cloud droplets in the  $0.3$  to  $20 \mu\text{m}$  range. Particle concentrations to  $10^3$  particles per  $\text{cm}^3$  at a throughput of  $1 \text{ cm}^3/\text{sec}$  can be accommodated. This chamber is also ideally suited to work in parallel for many of those experiments assigned to the expansion and static diffusion liquid chambers to provide supplementary or comparison data. A fully automated, slightly reconfigured CFD could then operate as a condensation nuclei characterizer.

**Static Diffusion Liquid Chamber (SDL)** - This chamber provides a controlled supersaturated environment above water freezing temperature and is used to obtain information concerning condensation nuclei numbers that exist in the natural environment. Its use in near zero gravity will be to determine the effects of gravity induced sedimentation and subsequently provide calibration factors which will permit the terrestrial useful range to be extended below the present 0.2 percent supersaturation limit. Additionally, the information gained through the comparison of 1-g and 0-g chambers will permit the design and establishment of a standardized SDL geometry that will then permit a reliable comparison of data from multiple units.

**Static Diffusion Ice Chamber (SDI)** - This chamber provides a controlled supersaturated environment below water freezing temperature and is used to study various phases of ice and ice/droplet interactions. The types of experiments involved permit this chamber to have larger dimensions than the CFD and SDL. Ice crystals and/or droplets are injected into the

chamber individually or in numbers and subsequent interactions are monitored while the environmental conditions are manipulated. Those experiments which require a saturated or supersaturated environment at below freezing temperatures are appropriate for this chamber. Ice nucleation, ice multiplication, ice crystal growth habits, riming and aggregation are representative experiment classes. Certain aspects of nucleation, growth, scavenging and charge separation are ideally suited to this type of chamber.

Expansion Chamber (E) - The prime feature of this chamber is that it most closely duplicates the natural cloud expansion processes through adiabatic expansion. This expansion influences the relative humidity which exists within the chamber and permits supersaturated and subfreezing conditions. Appropriate cooled chamber walls will permit, in zero gravity, experiment cycles which extend tens of minutes as occurs in clouds. Droplets and/or ice can be injected into the chamber singly or in numbers and the subsequent changes and interactions are monitored as an adiabatic cooling expansion occurs or as a heating compression takes place. The cycle can be repeated as required by the phenomena being studied. The main experiments suited for this chamber are those dealing with clouds of particles. This chamber provides the observation of multiple phenomena taking place simultaneously.

General Chamber (G) - Many of the dynamic experiments do not require supersaturated conditions within the chamber. On the other hand more access and control of the particles within the chamber are required. The General chamber satisfies the requirements of experiments represented by drop collision breakup and coalescence efficiency studies. In addition the controlled uniform, nonsaturated atmosphere is optimum for the study of some aspects of experiments like the saturation vapor pressure over supercooled water, collision induced freezing and riming/aggregation.

Earth Simulation (ES) - This special apparatus provides an enclosure to shield a rotating sphere from ambient thermal and air motions. Although a free-floating object is not required, the low-gravity environment permits

the desired radial gravity simulation by use of electrical fields. This equipment is suitable for the study of the circulation of planetary atmospheres and oceans.

### 2.3 RESEARCH RELATED EQUIPMENT

Evaluation of the detailed Experiment Class descriptions revealed significant commonality in the research tools used throughout the Experiment Program. The research tools were identified in three broad categories.

- Optical Detectors and Imaging Devices
- Droplet Generators
- Particle Detectors and Characterisers

It was further found that these research tools were used in different combinations dependent upon Equipment Class objectives. The CPL concept, therefore, has incorporated these research tools in modular units permitting their usage with the degree of flexibility required by such a dynamic area of research.

### 2.4 MISSION ASSESSMENT

The CPL goal is to be a general-purpose facility capable of accommodating the broadest range of experimentation. The experiments defined to date are envisioned to change or be modified by the results of on-going terrestrial research, cloud modeling, and field experimentation efforts. This goal was identified by realization that there exists sufficient microphysical research to be performed that requires or benefits by conduct on a manned orbital platform. The Experiment Class definitions were evaluated to quantify the Experiment Program size and therefore verify the initial assumption.

It must be realized that the quantification of the Experiment Program for the CPL must be extrapolated from the Experiment Class data provided by the scientific community. These definitions, although detailed to sufficient depth, can provide only coarse approximations of the total orbital experiment time required for their accomplishment. The individual experiment



events vary significantly in the time for their performance, the CPL operations between events are estimated, the "capability level" of the scientist-astronaut has been assumed and the many other aspects influencing experiment efficiency are not sufficiently well known to provide high confidence. The orbital experiment time, and hence mission assessment, is sufficiently accurate to substantiate and justify the general-purpose facility concept for the CPL.

The effective experiment observation time for each experiment class is shown in Figure 2-3. Only those events and operations concerned with microphysical research are included in the effective experiment observation time. The wide variation in duration has resulted from consideration of the primary and alternate cloud chamber usage, the parameters to be evaluated with the experiment class (Figure 2-4), the data gathering instrumentation variations and other pertinent experiment considerations. Both the range and the nominal event duration are shown. For the mission assessment the nominal duration was used as typical for the experiment class.

The experiment classes and the parameters of Figure 2-4 were established based on the experiment class definitions and consideration of CPL capability to provide parameter control. The significant quantity of parameters and their control individually and in combination is provided to satisfy the specified and envisioned desires of the scientific community users. The primary and secondary parameters are shown in the figure. The primary parameters identified are those that terrestrial researchers have identified as having major influence on the microphysical phenomena to be evaluated. The secondary parameters are those whose individual significance to the phenomena being studied is considered minor or must be deferred until the basic phenomena are sufficiently well understood. Combinations of secondary parameters can, however, be of significant importance and capability for their evaluation is therefore provided.

A nominal (or mean) number of variations for each parameter was established and is shown in Figure 2-4. The number of experiment observations was

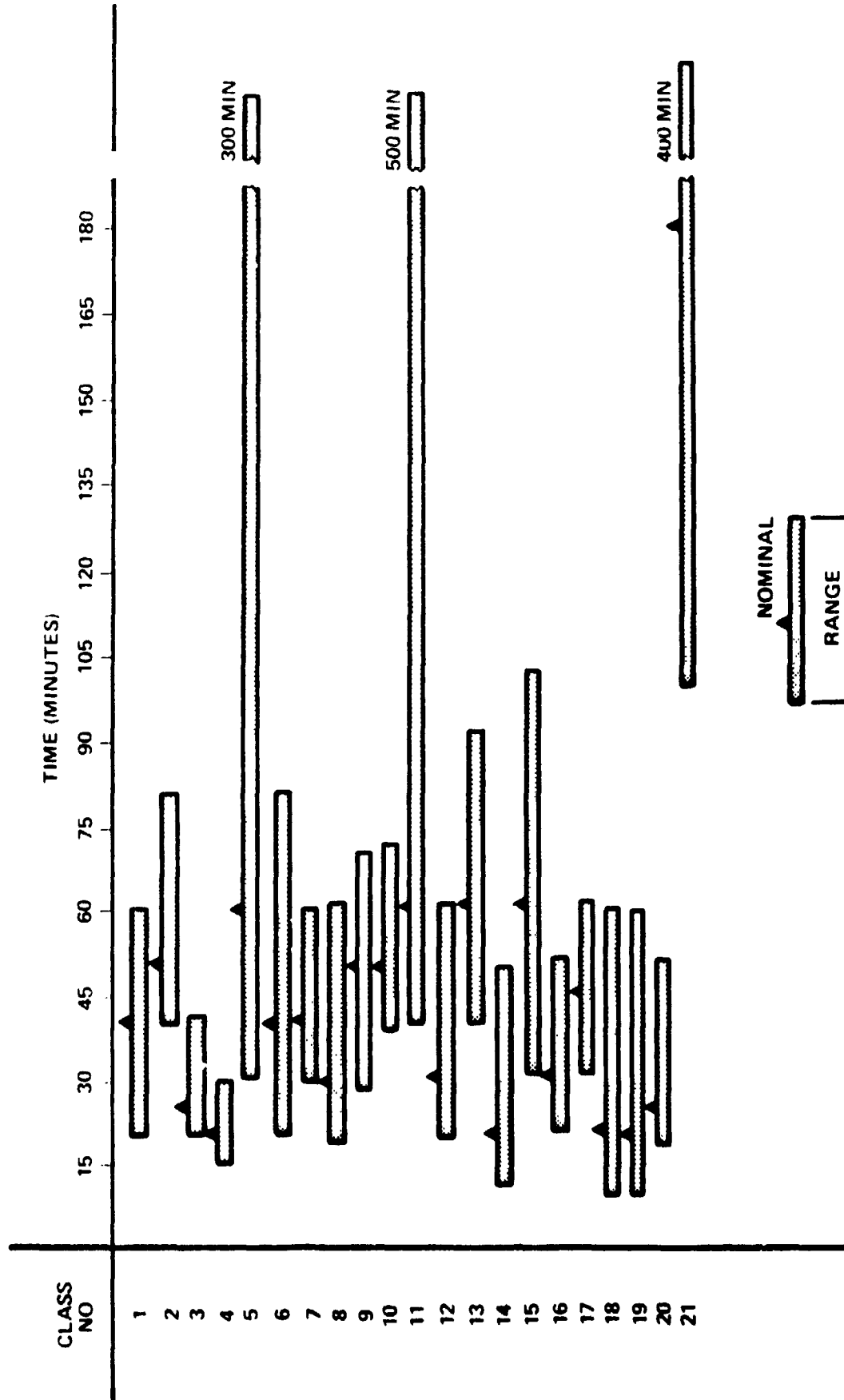


Figure 2-3. Effective Experiment Observation Times

CLASSES	PARAMETERS																				
	SIZE-NUCLEI	SIZE-DROPLET	TYPE	POLLUTANT	PRESSURE	TEMPERATURE	RELATIVE HUMIDITY	CHARGE	RATE OF COOLING	TIME	SOUND	ELECTRIC FIELD	NUCLEAR RADIATION	ADSORPTION	TURBULENCE	VENTILATION	OPTICAL	SHAPE	ORIENTATION	CONCENTRATION	VELOCITY
1. CONDENSATION NUCLEATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
2. ICE NUCLEATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
3. FREEZE SPLINTERING	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
4. CHARGE SEPARATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
5. ICE CRYSTAL GROWTH HABITS	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
6. SCAVENGING	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
7. RIMING AND AGGREGATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
8. DROPLET-ICE CLOUD INTERACTION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
9. HOMOGENEOUS NUCLEATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
10. COLLISION-INDUCED FREEZING	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
11. SATURATION VAPOR PRESSURE	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
12. ADIABATIC CLOUD EXPANSION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
13. ICE NUCLEI MEMORY	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
14. TERRESTRIAL EXPANSION CHAMBER EVALUATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
15. CONDENSATION NUCLEI MEMORY	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
16. NUCLEI MULTIPLICATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
17. DROPLET COLLISION BREAKUP	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
18. COALESCENCE EFFICIENCIES	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
19. TERRESTRIAL STATIC DIFFUSION CHAMBER EVALUATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
20. UNVENTILATED DROPLET DIFFUSION COEFFICIENTS	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
21. EARTH SIMULATION	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
MEAN NUMBER OF VARIATIONS	4	4	6	4	3	4	4	3	3	6	3	3	3	4	4	3	4	4	3	3	4

X - PRIMARY      O - SECONDARY

Figure 2-4. Experiment Parameter Requirements

established by addition of the parameter variations of the primary parameters only. It is possible to obtain a significantly large quantity of experiment observations by determining the factorial total of both primary and secondary parameters. This would, from the scientific standpoint, provide total "mapping" of the phenomena. The quantity of experiment observations specified for the CPL Experiment Program is therefore conservative (i.e., a much large experiment program can result).

The quantity of experiment missions is shown in Table 2-4. The nominal effective experiment observations times are multiplied by a factor of two to obtain total experiment event time. This 50 percent experiment efficiency factor was considered appropriate based on the feasibility study results and consideration of the operations to be conducted between events. This total experiment event time, the number of parameter variations and the quantity of event iterations were used to determine the number of experiment events and the total number of experiment hours (Table 2-4). By assuming 5 days of experiment operation (during a 7-day mission) and 8 hours of experimentation a day, the number of missions were calculated (40 hours of experimentation per mission).

The significance of the mission assessment identifying 138 missions lies not in the value specified but in the scope of the total research. This scope substantiates the rationale and goal of providing a general purpose facility for the conduct of cloud microphysics research. It should further be noted that the evolving Shuttle/Spacelab projects may revise the number of experiment hours per day and the number of days of on-orbit experimentation (growth to 30-day missions). The general-purpose CPL concept is viable with such change and growth and although the number of missions will vary the >5,000 hours of experimentation can be accommodated.

It should be noted that the Experiment Class 21 (Earth Simulation) was included in the Experiment Program because of its compatibility with the other experiment classes. This macroscale experiment class requires long

Table 2-4  
EXPERIMENT PROGRAM MISSION SUMMARY

Experiment Classes	Parameter Variations	Experiment Hours	Missions	
1 Condensation Nucleation	49	326	8	} Total 138
2 Ice Nucleation	52	424	11	
3 Ice Multiplication	23	384	10	
4 Charge Separation	32	320	8	
5 Ice Crystal Growth Habits	32	320	8	
6 Scavenging	33	160	4	
7 Riming and Aggregation	24	480	12	
8 Droplet-Ice Cloud Interactions	22	220	6	
9 Homogeneous Nucleation	43	326	8	
10 Collision Induced Freezing	25	226	6	
11 Saturation Vapor Pressure	21	420	11	
12 Adiabatic Cloud Expansion	29	176	5	
13 Ice Nuclei Memory	33	250	6	
14 Terrestrial Expansion Chamber Evaluation	18	180	5	
15 Condensation Nuclei Memory	25	68	2	
16 Nuclei Multiplication	24	80	2	
17 Drop Collision Breakup	29	132	3	
18 Coalescence Efficiencies	28	760	19	
19 Static Diffusion Chamber Evaluation	25	84	2	
20 Unventilated Droplet Diffusion Coefficients	18	60	2	
21 Earth Simulation	TBD	TBD	TBD	

experiment observation durations and has two primary parameter variations (Figures 2-3 and 2-4). The number of missions required for accomplishment of the objective requires additional evaluation but for planning purposes nine missions are envisioned.

## **2.5 PRIORITY MISSION RANKING**

As presently envisioned the CPL project will use two flight articles and accommodate a launch rate of 4 per year (maximum) equally distributed over the year (one every 3 months). Each flight article is to have a life expectancy of 10 years (20 missions) and capability for extended duration on-orbit experimentation (compatible with Shuttle/Spacelab growth). The total envisioned Experiment Program cannot therefore be accommodated in the first 10 years of flight\*.

Priority ranking is therefore necessary and evaluation of the priority mission requirements to total experiment program requirements is necessary.

The mission priority assessment (Table 2-5) was determined using the inputs from the scientific community. Each class was assessed for priority factor, applicability to zero gravity, and achievement ability. A ranking of A (highest), B (nominal), and C (lowest) was provided in each category. The individual scientific community rankings were summarized and factored using the numerical values shown in Table 2-6.

The mission priority ranking developed by the above approach is shown in Table 2-5. The ranking is not deemed to be absolute since only minor numerical total differences exist between many experiment classes. Furthermore, continuing terrestrial efforts will tend to change the priority assessment.

During the 10-year life of the CPL, 40 missions are planned. These missions are allocated (in quantity only, no mission flight order) in

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\*Project logic assumes technology advancement and subsystem modular replacement with advanced systems during the 10-year life. It was further assumed that subsequent research would be performed in an advanced CPL.

Table 2-5  
**EXPERIMENT MISSION PRIORITY RANKING**  
 (First 38 Missions)

Experiment Class Priority	Class No. and Title	Number of Missions	Assessment
1	5 Ice Crystal Growth Habits	3	<ul style="list-style-type: none"> <li>• Includes first 17 priority experiment classes</li> <li>• Utilizes all cloud chambers</li> <li>• Requires all laboratory subsystems</li> <li>• Potential reduction in parameter ranges and tolerances</li> </ul>
2	6 Scavenging	2	
3	18 Coalescence Efficiencies	3	
4	2 Ice Nucleation	3	
5	3 Ice Multiplication	3	
6	1 Condensation Nucleation	3	
7	20 Unventilated Droplet Diffusion Coefficients	2	
8	13 Ice Nuclei Memory	2	
9	4 Charge Separation	2	
10	12 Adiabatic Cloud Expansion	2	
11	16 Nuclei Multiplication	2	
12	19 Static Diffusion Chamber Evaluation	2	
13	17 Drop Collision Breakup	2	
14	14 Terrestrial Expansion Chamber Evaluation	3	
15	10 Collision Induced Freezing	2	
16	7 Riming and Aggregation	2	
17	8 Droplet-Ice Cloud Interactions	0	
18	9 Homogeneous Nucleation	0	
19	11 Saturation Vapor Pressure	0	
20	15 Condensation Nuclei Memory	0	
TBD	21 Earth Simulation	TBD	

Table 2-6  
PRIORITY ASSESSMENT NUMERICAL FACTORS

Ranking Value	Priority Factor	Applicability to Zero Gravity	Achievement Ability
A	64	25	11
B	54	18	7
C	43	11	3

Table 2-5. In general the number of missions allocated was established based on the total number of experiment hours for the experiment class. As shown, only 38 flights were allocated, leaving two mission flights for Experiment Class 21, Earth Simulation. Only four experiment classes (8, 9, 11, and 15) are not anticipated for conduct in the first 40 missions.

Experiment Class 21, Earth Simulation, was not prioritized in accordance with the previously described procedure. Since this experiment class involves macroscale phenomena evaluation, comparison and evaluation with microscale phenomena by microscale phenomena scientists was not deemed appropriate. This experiment class is deemed of significant importance and 2 missions of the first 40 missions have been allocated for its performance.

#### 2.6 PRIORITY MISSION SET - TOTAL EXPERIMENT PROGRAM REQUIREMENTS COMPARISON

Evaluation of the priority mission set to total experiment program requirements showed that only minor tolerance and range differences exist. All cloud chambers, droplet generators, particle detectors/characterizers and optical and imaging devices are required. The lack of significant requirements differences combined with the potential for changing priorities, the desire to maintain flexibility for the greatest range of research and the encouragement of total scientific community support resulted in the decision to formulate the CPL concept for the total experiment program.



**Appendix A**  
**CLASS I**  
**CONDENSATION NUCLEATION**

**A. 1 CONDENSATION NUCLEATION APPROACH 1**

**A. 1. 1 Introduction**

The topic of condensation nucleation has progressed to the point where there is some understanding of the process, as demonstrated by the fair successes attained in attempts to predict the concentration of droplets near the base of a convective cloud from measurements of the spectrum of critical supersaturations of cloud nuclei (Squires and Twomey, 1961; Jiusto, 1966; Twomey and Warner, 1967; Fitzgerald, 1972). Because of the difficulty of obtaining representative and related measurements in the free air, this kind of experiment can give only a very approximate confirmation of the existing theories of condensation. More detailed examination of the process must be carried out under controlled conditions.

Little has been done along these lines; Twomey (1965, 1971, 1972) has made determinations of the diffusion coefficient of cloud nuclei, and has carried out experiments which indicate that the majority of natural cloud nuclei have a degree of volatility similar to that of ammonium sulfate. Chodes, Warner and Gagin (1974) have recently made a determination of the sticking coefficient for condensing water vapor molecules, which was essentially in agreement with the generally favored value of Alty and Mackay (1935), but nothing is known of the variations in this coefficient in time and place.

**A. 1. 2 Objectives**

Readily identified objectives of experiments on condensation nucleation therefore include:

- A. An experimental check on the theory of cloud formation;
- B. Examination of the sizes and constitution of natural cloud nuclei;

- C. Determination of the kinetic effects associated with less than unity values of the sticking coefficient.

The highest priority would be A above, which would require the determination of a spectrum of critical supersaturations and the measurement of the droplet spectrum in an expansion chamber at the various stages of a slow expansion. It should be noted that in such an experiment, the "initial humidity" problem of the expansion chamber is not relevant, since it is not necessary to determine the supersaturation in the chamber by calculation; the only required output is the droplet spectrum as a function of  $\Delta p$ .

#### A.1.3 Justification and Applications

The importance of the nucleation of droplets which form a cloud arises from the fact that the microstructure of the cloud is determined at this point. It is well established by both theory and observation that continental clouds, in which the droplet concentrations are hundreds and sometimes thousands per  $\text{cm}^3$ , behave quite differently from maritime clouds with 50-100 droplets per  $\text{cm}^3$ . Maritime clouds usually form rain once they exceed about 2 km in depth by the warm rain process, whereas continental clouds are virtually restricted to forming precipitation by the cold, or Wegener-Bergeron process.

One consequence of this is that in summer, when convective clouds are the dominant type over the continent (as in Colorado) the clouds must grow quite deep before they can precipitate. By this time, they are often large enough to be thunderstorms, or to form hail.

Even in clouds in which the ice phase plays a role, there is considerable evidence both from cloud observation (e. g. Mossop, 1971) and from laboratory work by Hallett and Mossop (1974) that the observed ice particle concentrations, in maritime clouds, are strongly influenced by a multiplication process which probably will not work efficiently in continental clouds because the droplets are too small. The implications of this with respect to the electrical effects in thunderstorms and the generation of hail are yet to be determined, but seem likely to be of crucial importance.

The rationale for cloud seeding procedures has always rested on a rather meager foundation. Clouds are seeded in the belief that by catalyzing, more ice particles are formed and precipitation can be increased, or clouds can be influenced in some other desirable way. It is often assumed, without knowledge of the actual ice particle concentration, or an understanding of how it can be predicted, that more ice particles will be beneficial; it is obvious that if the natural concentration is high, seeding may not produce any effect, or even a deleterious one.

The details of the condensation process are therefore one of the keys to rational control of weather modification procedures, and in addition, when well understood, may conceivably make available a totally new method for controlling the behavior of clouds through modification of the cloud-forming process itself.

#### A.1.4 Terrestrial Limitations

The limitations of aerosol and condensation nucleation experimentation in 1-g arise from sedimentation of particles and convection currents caused by temperature differences. The development of the static diffusion chamber has been difficult because of the fall-out of nucleated drops.

The use of diffusion chambers in meteorology arose from the need to establish and maintain quite low supersaturations at a designed level of order 1 percent for periods of several seconds — a task for which expansion chambers are unsuited, even under zero-g conditions because of the initial value problem (R. H. known to 0.05 percent or so). In a diffusion chamber, instead of trying to minimize the disturbing influence of the walls on an adiabatic gas process, one uses the influence of the walls to establish the desired conditions. The above constraints are used to establish the dimensions of diffusion chambers to a very large degree. The depth of a diffusion chamber must be kept small for the following reasons:

- A. To obtain large diffusive fluxes, in order to have good control of the supersaturation in the center of the chamber despite the presence of growing particles;
- B. To suppress convection and side-wall effects;

C. To keep the set-up time required for the establishment of the desired supersaturation small.

The requirement to keep the depth small has the result that at 1-g, fall-out is a major problem in the use of diffusion chambers, and limits the extent to which advantage can be taken of their ability to maintain a constant supersaturation(s) for many seconds, despite the growth of particles. At constant supersaturation the droplet grows roughly according to  $dr^2/dt = 2 \times 10^{-8} S$ ; it falls according to  $z = 10^6 r^2$ . Hence,  $z = 2 \times 10^{-2} S$ , and the distance fallen after time  $t$  is  $10^{-2} S t^2$ , or  $10^{14} r^4 / 4 S$  cm. Thus with any given geometry, the problems of keeping the droplets in the illuminated region until they grow to detectable sizes become worst as  $S$  decreases.

In some degree the fall-out problem difficulty has been mitigated in the CFD chamber by transporting the drops out of the diffusion chamber into a separate optical detector, in which positive droplet detection can be made at smaller sizes, and by the introduction of vertical-walled chambers (Sinnarwalla and Alofs, 1973; Hudson and Squires, 1973, 1974). The N. R. L. group is working on an isothermal vertical-walled chamber in which the supersaturation is generated chemically, as in the early work of Twomey (1959). These attempts are still under development, but will certainly continue to be limited by the effects of sedimentation and convection.

The analysis given in Attachment A of this Approach 1 indicates that in zero-g, the depletion of a stored aerosol may be reduced from its terrestrial value of about 5 percent per hour for cloud nuclei to about 1 percent per hour or less, chiefly as a result of the reduction in the velocity of convection currents.

#### A. 1.5 The Opportunity of the Zero-g Laboratory

The advantages of "zero-g" for cloud chamber work stem from the elimination (or reduction) of both sedimentation of particles and of convection currents.

These effects will be discussed in connection with aerosol storage and chamber operation.

#### A. 1. 5. 1 Aerosol Storage

In the case of aerosols generated on board, it may prove advantageous to generate a volume of aerosol and store it for successive experiments of various kinds. An example of this would be, to measure the spectrum of critical supersaturations, and to compare this with a size spectrum in order to check the validity of standard Köhler theory which relates the critical supersaturation ( $S_c$ ) to the mass of a soluble particle of known constitution. Another would be to form a cloud by adiabatic expansion and to compare the resulting spectrum of droplet sizes with that predicted from the spectrum of  $S_c$  by computation, following Howell (1949), Twomey (1959), Fitzgerald (1972) or otherwise. Even in the area of particle size measurement, it may be advantageous to use the rather slow diffusion battery technique in order to check the validity of electrostatic measurements. All such experiments will require periods of an hour or more, and it may be possible to achieve greater aerosol stability by storing a volume from which samples are drawn than by other methods using a continuous flow generator.

Samples of the Earth's atmosphere may be studied to great advantage in zero-g, since more accurate and reliable results can be obtained than in terrestrial experiments. Since the natural aerosol is extremely complex, consisting of a mixture of particles of diverse origins and of mixed constitutions, no experimentation on artificial aerosols can replace natural samples. Experimentation with Earth samples will certainly require storage for periods of at least half a day, even if an uncontaminated sample can be drawn into an already-stowed container shortly prior to launch.

In Attachment A, a discussion is given of the particle losses which occur when an aerosol is stored in a container with a typical linear dimension,  $L$ , of 50 cm. In a terrestrial laboratory, the observed loss rate of cloud nuclei is typically about 5 percent per hour. It is shown in Attachment A that the various mechanisms of removal (Brownian diffusion to the walls, thermophoresis, coagulation and sedimentation) are quite inadequate to explain such a loss rate, unless account is taken of the convection currents which ordinarily must be present as a result of small temperature differences across the container (taken to be in the range  $\delta T = 0.1$  to  $0.01^\circ\text{C}$ ). When account is taken of the effects of such currents, the loss by diffusion is very greatly accelerated, and appears capable of explaining losses of the order of 3 percent per hour.

The theory indicates that the loss rate due to diffusion is proportional to  $(g\delta T/L)^{1/4}$ . Thus, if in a zero-g laboratory,  $g$  is reduced to  $10^{-5}$  of terrestrial  $g$ , the diffusional loss rate would be reduced by a factor of about 17, to values of about 0.3 percent per hour. Some improvement could be achieved by enclosing the container in an outer shell to make it more isothermal, but no practicable increase in container dimensions would contribute significantly.

The mechanisms other than diffusion which would remain effective in zero-g (thermophoresis and coagulation) appear to be capable of causing losses of about 0.3 percent per hour, so that the total loss rate would be less than 1 percent per hour. By suitably adjusting the total aerosol size spectrum, it would be possible to balance the elimination of cloud nuclei by coagulation against the formation of new cloud nuclei by the coagulation of smaller particles. Alternatively, for many purposes, it may be appropriate to reduce the coagulation loss by diluting the sample with particle free air; in that case, the total could be essentially that due to diffusion, or about 0.3 percent per hour. Thus a sample stored for half a day would be depleted by only about 10 percent or less, which for many purposes may be quite tolerable.

In the case of Earth samples, it would be desirable to introduce a local sample (e.g. a maritime aerosol at Cape Kennedy) into an already-stored container at a late stage during the preparations for launch. The effect of high  $g$  during injection into orbit on a stored aerosol is likely to be minor. Thus in the case of sedimentation, 3  $g$  lasting for 200 seconds would remove as many particles as 1  $g$  would in 600 seconds, that is about 0.1 percent, while the loss caused by diffusion would be about  $3^{1/4}$  times that which would occur on Earth in the same time period, i.e. about  $(1.3 \times 200/3600 \times 5) = 3$  percent.

#### A.1.5.2 Chamber Operation

Sedimentation of the droplets which grow in the supersaturated environment of a cloud chamber is, of course, a very serious limitation. For diffusion chambers with horizontal plates, it limits the usable range of supersaturations to above 0.2% (Twomey, 1967; Squires, 1972), both in static and continuous chambers. When the plates are vertical as in the chamber of Sinnarwalla

and Alofs (1973), or a variant of the Hudson and Squires (1973, 1974) chamber a convective cell is superimposed in the general flow, and this considerably complicates the operation of the chamber. A chamber in zero-g would be largely free of these complexities and would be limited only by the transport of particles as a result of phoretic effects.

In an expansion chamber, the reduction in zero-g conditions of the convection currents due to differences between the wall temperature and that of the bulk gas is of the greatest importance. Following the analysis given in Attachment A, the velocity of such currents will be of the order of  $(0.1 g \kappa L \delta T / \nu T)^{1/2}$  where L is the characteristic dimension of the chamber. Thus, if  $\delta T$  can be kept as small as  $0.1^\circ \text{C}$ , and g is reduced below its terrestrial value by a factor of  $10^5$ , if L is taken to be 50 cm, a characteristic velocity will be of order  $10^{-2} \text{ cm sec}^{-1}$ , and the movement of the air as a result of convection will not produce significant transport of either heat or water vapor from the neighborhood of the walls to the central part of the chamber in periods less than several hundreds of seconds. Provided the pressure within the chamber is monitored during the expansion (or held constant) so that the interpretation of the experiment is not affected by volumetric changes due to the thermal influence of the walls on the immediately neighboring air, the only cause of departures from adiabatic conditions would be thermal conduction and vapor diffusion. In a chamber of radius  $a = 25 \text{ cm}$ , at time  $t = 100 \text{ sec}$ , the departure from adiabatic of the central temperature  $\Delta T$  is only a small fraction of the wall temperature,  $\delta T$ .

Following the treatment of diffusion in a sphere of radius a cm given in Attachment A it is seen that a departure of the wall temperature by  $\delta T^\circ \text{C}$  from the control temperature will produce a disturbance of that temperature of  $\Delta T$  at time t, where:

$$\frac{\Delta T}{\delta T} \approx \frac{a}{(\kappa \pi t)^{1/2}} \exp \frac{-a^2}{4 \kappa t}$$

$$\approx 3 \exp (-8) \approx 10^{-3} \text{ at } t = 100 \text{ sec.}$$

The disturbance in the water vapor concentration will also be negligible, since the diffusivity of water vapor in air is similar to that of heat.

It is evident that experiments lasting a few hundred seconds would be possible, the acceptable duration being limited primarily by convective currents, the velocities of which would be proportional to  $(g\delta T)^{1/2}$ .

#### A.1.6 Quantification

The first diffusion chamber used in meteorology was that of Wieland (1956), followed by Twomey (1959). In the early 1960's Twomey developed what is still the best version of the thermal chamber; others also worked with static thermal chambers, but great difficulties were met, basically because of droplet sedimentation. It may be safely said that our present knowledge of the properties of cloud nuclei from diffusion chamber experiments would have been achieved in a zero-g environment, in five years, instead of in the eighteen years which have elapsed since the first experiment. Indeed, since the need for such knowledge was clearly stated by Squires (1952), and was no doubt realized by others still earlier, the "acceleration ratio" would probably have been at least 4:1.

#### A.1.7 Approach

The approach to be used to investigate the three major experimental areas is presented below:

##### A.1.7.1 Check on the Theory of Cloud Formation

The formation of a convective cloud would be modelled in an expansion chamber with "synchronous" wall temperature, and prior to the modeling, the spectrum of critical supersaturations would have been determined.

An experiment of this kind was attempted by Gunn and Phillips (1957), using a very large sphere without wall temperature control. According to the laminar-flow theory of Appendix A, the convective velocities generated by a inhomogeneity of 5°C would have been of the order of a meter a second, but such flows would certainly have been turbulent, and may have been even faster, resulting in a far-from-adiabatic condition in the bulk gas.



#### A.1.7.2 Sizes and Constitution of Cloud Nuclei

Work in this area has been carried out by Twomey (1972) and by Hoppel, Dinger and Ruskin (1973) using similar equipment. The volatility of nuclei was determined by passing the sample through a heated quartz tube, and their size by the use of diffusion batteries. In Twomey's later work (1972), these are extremely compact devices, the batteries themselves consisting of nuclepore filters. Electrostatic methods could also be used to help determine the particle size spectrum.

Basic investigations would be made using an artificial soluble-salt aerosol to check that the spectrum of critical supersaturations was in agreement with the spectrum of particle sizes; but the most important work would employ Earth samples, the objective being to determine to what extent the natural nuclei consist of soluble and insoluble components.

#### A.1.7.3 Kinetic Effects

Kinetic effects experiments are valid only when the droplets are grown with accurately known chamber conditions and the resulting droplet sizes are determined as a function of time. Experiments by Chodes, Warner and Gagin (1972) have indeed tended to confirm the earlier work of Alty and Mackay (1935), but it must be acknowledged that these experiments are of great difficulty, since they involve doubly differentiating the raw data — that is, the vertical coordinate of the droplet being studied.

Other approaches to the same problem are possible, such as that outlined by Rogers and Squires (1974), and all these methods would be simplified and made more certain in a zero-g environment.

#### A.1.7.4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	
Type	11
Pollutant	4

<u>Parameters</u>	<u>Variations</u>
Pressure	1
Temperature	4
Relative humidity	
Charge	
Rate of cooling	4
Time	2
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	3
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	3
Kinetic energy	
Gases	
Spin rate	

#### A. 1. 8 Detailed Requirements

Diffusion chambers require a means of fixing the temperature of the plates to within  $0.05^{\circ}\text{C}$  over an area which may range from less than  $100\text{ cm}^2$  for small static chambers up to over  $1000\text{ cm}^2$  for continuous flow chambers.

Arrangements are also needed for keeping the plates thoroughly wet. The usual solution is to use filter paper supplied by a flow of water, which exceeds the evaporation losses, so that drainage is also necessary.

Unwanted pressure changes must be avoided, since they could modify the supersaturation between the plates. This presents no problem in a static chamber, because each sample can be isolated in the chamber from environmental pressure changes, but in a CFD chamber this will be more difficult. Since the diffusive time constant is typically about 1/2 sec, it is necessary that significant pressure changes (of order 0.01 mm, corresponding to about  $10^{-2}$  percent change in the supersaturation) should occur only over periods of time long compared with 1/2 sec. The maximum rate of change of pressure should therefore be less than a few times  $10^{-3}$  mm sec<sup>-1</sup>.

The handling of aerosols can present problems because of the possibility of diffusive losses in tubing. Some devices, such as SD chambers, require the sample to be forced in from a lung or other collapsible container, or drawn in from a bag at ambient pressure, and this can be done at a high volume flow rate, which minimizes diffusive losses. A CFD chamber on the other hand requires a steady flow of the order of 1 cm<sup>3</sup> sec<sup>-1</sup>, and tubing length from a reservoir (or larger flow) must be kept very short – of the order of 10 cm.

A prime need for the employment of a CFD chamber would be the development of an optical counter which is well calibrated for water droplets and is free from the ambiguity which is present in several present day optical particle sensors. For the measurement of droplets in the 1-2 micron range which is required for nucleus counters and for the determination of kinetic effects, the choice at present lies between the method of Chodes, Warner and Gagin (1974) in which size is deduced from fall speed in air which is presumed to be stationary, or optically determining the size, which essentially implies that the droplet be transported from the region of formation to a separate sensing device.

#### A.1.9 Procedure

General activity details are given below followed by a representative activity timeline. The event sequence and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations of this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient.

It would be desirable to have two or even three identical CFD chambers in simultaneous operation as is currently done in the laboratory. One percent agreement between counters has already been achieved. Multiple counters would not only have advantages in determining fundamental parameters but would also greatly speed the progress of experiments.

All equipment must be turned on and allowed to warm up. It takes about one-half hour for the Royco electronics to stabilize so that the secondary calibration can be checked and set. (See Figure A-1). The sizing channels can now be set and they can be counted on to remain stable for the entire mission. Of course, if different sizes are wanted they will have to be reset. The counting interval should also be checked and set (1 min.). During the initial warm-up, the water feed bottle should be filled, the drain emptied and the water feed to the plates set so that they will stay wet. The Royco pump should be run as much of the time as possible so it, too, can stabilize (~40 min.). A preliminary pressure and flow setting can be made during warm-up.

The plate temperatures will also have to have time to stabilize, and the time required to reach equilibrium will depend on how much they differ from ambient temperature and if a particular  $\Delta T$  is desired. Desired temperatures will have to be re-set as they are approached.

When all parameters are set as they should be set, it is necessary to run for a few minutes with the sample inlet closed to make sure that the count is zero or to at least determine the background count. Then about four 1-minute runs are taken and the number of counts in each channel is averaged. The calculations can be done while the next set of data is being taken. Then the main flow  $F$  is changed while  $\Delta p$  is held constant and another set of four 1-minute runs is taken. At least three  $F$ -values are needed for a plateau. Experience can usually speed this process.

It is also desirable to vary  $F_1$  and  $F_2$  so that the position of the sample between the plates is optimal. This is usually not very critical and three

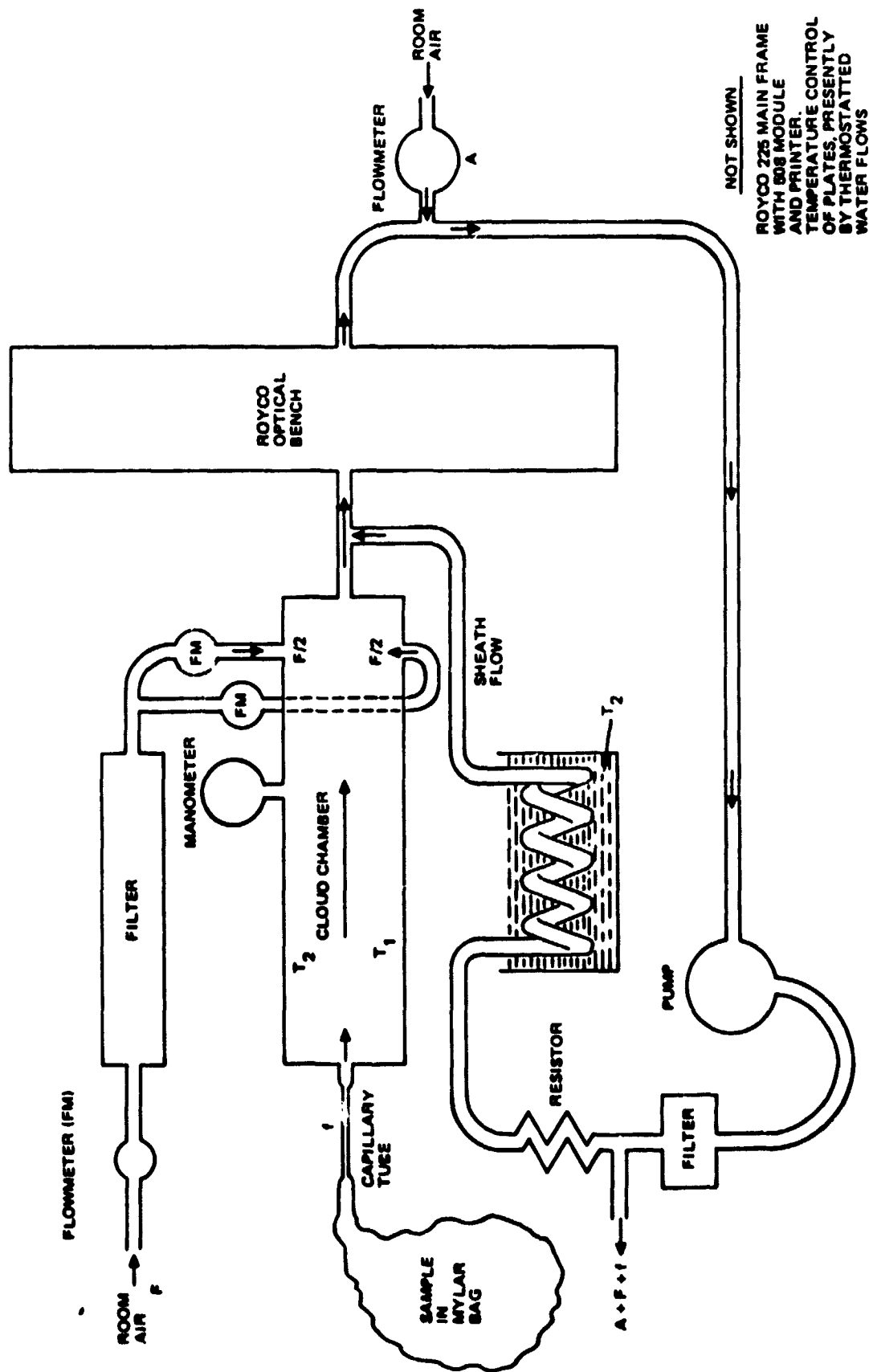


Figure A-1. CFD Layout

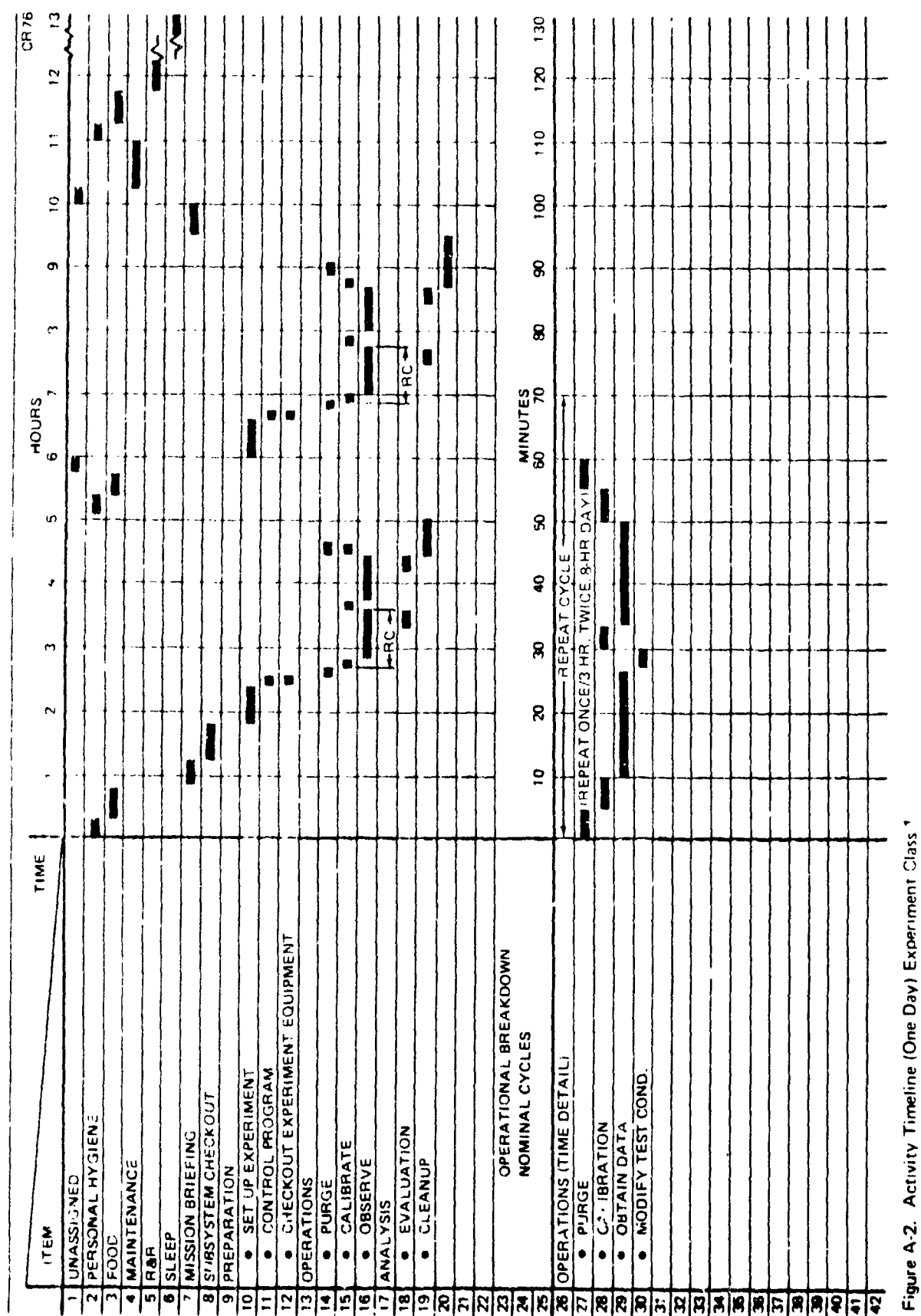
points should suffice. This would be done in the same manner as above. Change  $F_1$  and  $F_2$  while holding  $F$  and  $\Delta p$  constant.

Since the sample decays in its container, it is necessary to normalize the counter. If only one cloud chamber can be used this is done by returning to the original value of the parameter being varied ( $F$  or  $F_2/F$ ) then drawing a decay curve, and interpolating the equivalent values for the data points taken during the time in between. Sample decay is estimated to be  $\sim 1$  percent per hour (Attachment A). If two chambers are available, both should be set to the same  $\Delta T$  and all parameters can remain fixed in one chamber (they can be set at more or less optimum plateau values by experience). At the end of each set of experiments, a graph of  $N$  vs.  $F$  or  $N$  vs.  $F_2/F$  should be drawn for each size range; also a graph of  $N$  vs.  $d$  for each  $F$  and  $F_2/F$  to obtain a size plateau.

To describe the experimental procedure any further depends on the particular objective which the experimenter has in mind. A typical experiment is to determine  $k$  in  $N = CS^k$ . In this case  $\Delta T$  would have to be changed and the whole procedure repeated ( $F$ -plateau,  $F_2/F$  plateau, and size plateau). However, experience can cut down on the length of time this takes. At least three values of  $\Delta T$  are necessary to determine  $k$ , more would be desirable. If only one chamber is used, it would be desirable, if not necessary, to return to the original value of  $\Delta T$  in order to normalize for decay. Obviously two or even more chambers would make a  $k$  determination easier. Of course, this can be done in zero-g as easily as in 1-g. Better plate temperature controllers would decrease the time for changing temperatures and this would be most desirable. A detail activity procedure is given next followed by a timeline in Figure A-2.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Turn on all equipment	1 - 2
● Warmup time  During this time $\Delta P$ and flows can be preliminary set. Water feed tank can be filled and water flow to plates set.	30
● Check Royco secondary calibration	3
● Set size ranges on 508 module	5
● Set timing interval on 508	5
● Make final plate temperature settings	10
● Make final $\Delta P$ , $F$ , $F_2/F$ settings	3
● Determine background count	5
● Make four one-minute runs at these settings.	5
● Change $F$ while holding all other parameters constant	2
● Make four one-minute runs at this new $F$	5
● Change $F$ at least once more and repeat four one-minute runs for each value of $F$	15
● Change the value of $F_2/F$ for about two of the optimum $F$ values. Make four one-minute runs at these settings.	30
● Average counts for each set of runs. Normalize (see text). Draw graphs of $N$ vs $F$ and $N$ vs $F_2/F$ .	20
● Change plate temperatures	10-25
● Repeat steps above for new $\Delta T$ .	175





#### A.1.10 References

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## Approach 1 - Attachment A

# THE DEPLETION OF A STORED AEROSOL IN THE PRESENCE OF CONVECTION CURRENTS

## INTRODUCTION

Some experiments with aerosols require repeated measurements over a period of time of the order of an hour. Examples are the determination of a spectrum of critical supersaturations of cloud nuclei, or of a size spectrum by means of diffusion batteries; in the latter case, the inversion of the data is complicated by the fact that the aerosol which is being studied is changing while the successive runs are being made through diffusion batteries with different permittivities.

Aerosols are frequently stored for these purposes in Mylar bags which are internally aluminized to form a Faraday cage, in order to avoid electrostatic removal of charged particles. Such bags usually have a capacity of the order of 300 liters, and when filled have minimum linear dimensions of about 50 cm. In such a container, the particle count may be depleted by several mechanisms. The discussion will be restricted to cloud nuclei, that is, condensation nuclei with critical supersaturations less than 1 percent. Even if these particles were pure soluble salts, they must have radii exceeding  $10^{-6}$  cm, corresponding to a diffusion coefficient of less than  $1.4 \times 10^{-4} \text{ cm}^2 \text{ sec}^{-1}$ . In fact, their average diffusion coefficient is around  $10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ , corresponding to a radius of about  $3 \times 10^{-6}$  cm. It will be assumed here that particles larger than  $r = 2 \times 10^{-6}$  cm ( $D = 3.6 \times 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ ) are cloud nuclei and vice versa. Observation shows that in a container of the type described, the loss rate of cloud nuclei under ordinary laboratory conditions is about 5 percent per hour.

## EXPECTED LOSS RATE FROM STILL AIR

In this section, it will be shown that the loss rate of cloud nuclei which is observed cannot be explained without taking into account the convective stirring motions which must arise from minor temperature differences.

### A. Loss by Diffusion

If the air in the container is completely still, the diffusive loss of particles is easily shown to be negligible. For a sphere of radius  $a$  cm, initially filled with an aerosol of concentration  $N$   $\text{cm}^{-3}$  and diffusivity  $D$   $\text{cm}^2 \text{sec}^{-1}$ , the concentration at the center is given by Carslaw and Jaeger (1959) as a function of time by:

$$\frac{N - N_c(t)}{N} = \frac{a}{(\pi Dt)^{1/2}} \sum_{n=0}^{\infty} \exp \left[ - \frac{(2n+1)^2 a^2}{4Dt} \right]$$

The infinite series is dominated by the expansion of  $z/1-z^8$  where  $z = \exp(-a^2/4Dt)$ , so that the proportional decrease in the central concentration at time  $t$  is  $N - N_c(t)/N$  is less than

$$\frac{a}{(\pi Dt)^{1/2}} \frac{z}{(1-z^8)}$$

For  $a = 25$  cm,  $D = 3.6 \times 10^{-5} \text{ cm}^2 \text{sec}^{-1}$ ,  $t = 3.6 \times 10^3 \text{ sec}$ ,  $z \sim \exp(-1205)$ , so that

$$\begin{aligned} \frac{N - N_c(t)}{N} &\sim \frac{a z}{(\pi Dt)^{1/2}} \\ &\sim 40 \exp(-1205) \\ &\sim 10^{-516} \end{aligned}$$

Clearly, the effect of diffusion in a container with a "diameter" of 50 cm in which the air remains quite still is totally negligible. It is of course unrealistic to assume that the air in the container will remain stationary for periods of the order of hours, and a rough theory developed below indicates that diffusive losses would be much larger than predicted above, as a result of convective motions caused by minor temperature differences, which under ordinary laboratory conditions would probably be of the order of  $0.1^\circ \text{C}$ , or perhaps as small as  $0.01^\circ \text{C}$ .

B. Thermophoresis

The effects of thermophoretic transport of particles as a result of a temperature gradient likely to occur in a laboratory are small. If it is assumed that the container air is quite stationary, the effect of a temperature gradient of even  $0.01^\circ\text{C}$  over 50 cm is to produce a velocity of only about  $5 \times 10^{-7} \text{ cm sec}^{-1}$  (Goldsmith and May, 1966) so that after an hour in air at rest, only about  $4 \times 10^{-3}$  percent of the particles would be removed.

In fact, in the presence of a convective cell, the resulting particle motion between the region of maximum velocity and the wall would be towards the wall in the descending branch and away from it in the the ascending branch, and these two motions would tend to cancel each other.

C. Effect of Coagulation

Coagulation will also act to change the particle spectrum; some cloud nuclei will be eliminated by coagulation among themselves, and others formed by the coagulation of smaller particles. The elimination process itself is quite slow, proceeding at a rate which, because of kinetic effects, is less than  $8\pi r D n^2$  particles per second, i. e. a proportional decrease of  $8\pi r D n$  per second, or  $9 \times 10^6 r D n$  percent per hour. Since  $r D$  is a decreasing function of  $r$ , it will be conservative to take  $r = 2 \times 10^{-6} \text{ cm}$ ,  $D = 3.6 \times 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ . Taking  $n = 500 \text{ cm}^{-3}$ , a typical value for cloud nuclei in a continental aerosol, this indicates a decrease of less than 0.3 percent per hour. This calculation ignores the polydisperse nature of the aerosol, which tends to increase the coagulation rate, but on the other hand the decrease in cloud nucleus concentrations will tend to be cancelled by the formation of new cloud nuclei by the coagulation of smaller particles, and indeed in the case of polluted air with a very high Aitken count, coagulation among many small particles may lead to an increase in the cloud nucleus concentration. The calculations of Junge (1969) on the evolution of a continental aerosol distribution indicate, over periods of the order of  $10^4 \text{ sec}$ , essentially no change in the total number larger than  $r = 2 \times 10^{-6} \text{ cm}$ , corresponding to the range of sizes of cloud nuclei.

#### D. Particle Loss by Sedimentation

Another cause of particle loss is sedimentation. If the air in the container were completely at rest, the number flux of particles in the range  $r_1$  up to  $r_2$  through any horizontal plane would be simply  $\int_{r_1}^{r_2} V_g dN$ , where  $N$  is the cumulative concentration. For sufficiently large particles, this formula is valid not only in the bulk gas, but also at a horizontal wall surface, but in the case of sufficiently small particles, it is not valid to assume that their concentration very close to the wall is the same as in the bulk gas, since that concentration already is much reduced by Brownian diffusion. However, sedimentation is an effective process throughout most of the range of sizes corresponding to cloud nuclei, and in the present discussion it will be assumed that the function  $N$  is everywhere the same.

In order to evaluate the integral  $\int V_g dN$ , it will be assumed that the size distribution follows Junge's law,  $dN/d\ln r = \alpha r^{-3}$  from  $r = r_2$  (the maximum size) down to  $r = r_1 = 5 \times 10^{-6}$  cm, and that from  $r = r_1$  down to the lower limit of cloud nucleus sizes  $r_m$  ( $\sim 2 \times 10^{-6}$  cm),  $dN/d\ln r$  is a constant. The total count of particles from the maximum radius down to  $r_m$  is then:

$$\begin{aligned} \int dN &= \int_{r_1}^{r_2} dN + \int_{r_m}^{r_1} dN \\ &= \int_{r_1}^{r_2} \alpha r^{-3} d\ln r + \left( \frac{dN}{d\ln r} \right)_{r=r_1} \ln \frac{r_1}{r_m} \\ &= \frac{\alpha}{3} (r_1^{-3} - r_2^{-3}) + \alpha r_1^{-3} \ln \frac{r_1}{r_m} \\ &\approx \alpha r_1^{-3} \left( \frac{1}{3} + \ln \frac{r_1}{r_m} \right) \end{aligned}$$

• since  $r_1^{-3} \gg r_2^{-3}$

Taking  $r_1 = 5 \times 10^{-6}$  cm,  $r_m = 2 \times 10^{-6}$  cm, this implies that the total cloud nucleus concentration ( $n$ ) is close to  $1.25 \alpha r_1^{-3}$ . Hence  $\alpha = 0.8 n r_1^3$ .

The sedimenting flux through unit area of a horizontal plane is

$$\int_{r_m}^{r_2} V_s dN = \int_{r_1}^{r_2} V_s dN + \int_{r_m}^{r_1} V_s dN.$$

The fall velocity,  $V_s$  is given by:

$$V_s = \frac{4}{3} \pi r^3 \rho_p g B$$

where  $\rho_p$  is the density of the particles and  $B$  their mobility. Thus,

$$\int_{r_1}^{r_2} V_s dN = \frac{4}{3} \pi \rho_p g \alpha \int_{r_1}^{r_2} r^{-1} B dr$$

The empirical formula due to Knudsen and Weber (1911) and Millikan (1923) is:

$$B = \left[ 1 + \frac{A l}{r} + \frac{Q l}{r} e^{-b r / l} \right] / 6 \pi \eta r$$

where  $A \approx 1$ , and  $l$  is the mean free path of air molecules. The value of  $Q$  and  $b$  have been estimated from experiments with various aerosols, and depend somewhat on the constitution of the particles. Taking the results of Millikan (loc. cit.) for oil droplets in air,  $Q \approx 0.3$ ,  $B \approx 1.25$ .

This gives the sedimenting number of flux as:

$$\int_{r_1}^{r_2} V_s dN = \frac{2}{9} \frac{\rho_p g \alpha}{\eta} \int_{r_1}^{r_2} (r^{-2} + A l r^{-3} + Q l r^{-3} e^{-b r / l}) dr$$

$$= \frac{2\rho_p g \alpha}{9\eta} \left| -r^{-1} - \frac{A\ell r^{-2}}{2} + \frac{Q\ell}{2} \{ -r^{-2} e^{-pr} + pr^{-1} e^{-pr} + p^2 \text{Ei}(-pr) \} \right| \int_{r_1}^{r_2}$$

where  $p = \ell/\ell$ .

Putting  $r_1 = 5 \times 10^{-6}$ ,  $r_2 = 5 \times 10^{-4}$ ,  $p = 2 \times 10^5$ , since  $\text{Ei}(-1) \approx -0.2$ , it results that all terms are small compared with the first two, and we may write, approximately:

$$\int_{r_1}^{r_2} V_s dN = \frac{2\rho_p g \alpha}{9\eta} \left| r_1^{-1} + \frac{A\ell}{2} r_1^{-2} \right|$$

Also, since in the range  $r_m$  to  $r_1$ , it is assumed that

$$\frac{dN}{d\ell n r} = \alpha r_1^{-3},$$

$$\begin{aligned} \int_{r_m}^{r_1} V_s dN &= \frac{2\rho_p g \alpha r_1^{-3}}{9\eta} \int_{r_m}^{r_1} (r + A\ell + Q\ell e^{-Pr}) dr \\ &= \frac{2\rho_p g \alpha r_1^{-3}}{9} \left[ \frac{1}{2} r^2 + A\ell r - \frac{Q\ell}{p} e^{-pr} \right]_{r_m}^{r_1} \end{aligned}$$

Substituting  $\alpha = 0.8 n r_1^3$  and, later, putting  $\rho_p = 1.5 \text{ g cm}^{-3}$ ,

$$\begin{aligned} \int_{r_m}^{r_2} V_s dN &= \frac{1.6\rho_p g n}{9\eta} \left| r_1^2 + \frac{A\ell r}{2} + \frac{1}{2} (r_1^2 - r_m^2) \right. \\ &\quad \left. + A\ell (r_1 - r_m) - \frac{Q\ell}{p} (e^{-Pr_1} - e^{-Pr_m}) \right| \\ &\approx 1.5 \times 10^6 n \left\{ \frac{3}{2} r_1^2 - \frac{1}{2} r_m^2 + A\ell \left( \frac{3}{2} r_1 - r_m \right) \right\} \end{aligned}$$

$$- \frac{Ql}{p} (e^{-pr_l} - e^{-pr_m})$$

$$\approx 10^{-4} n$$

Thus, in a container  $h$  cm deep, the proportional rate of loss of cloud nuclei due to sedimentation from still air is  $10^{-4}/h$  per second, or about 0.7 percent per hour for  $h = 50$  cm.

#### E. Summary of Expected Losses with Still Air

In the case of a Faraday-cage type container with characteristic linear dimensions of 50 cm, the expected loss rate per hour of cloud nuclei, assuming that the air in the container remains quite stationary, is summarized below:

Diffusion to walls	$\approx 0$ percent
Thermophoresis	$< 0.01$ percent
Coagulation	$< 0.3$ percent
Sedimentation	$< 0.7$ percent
Total loss rate per hour	$< 1.0$ percent

Since the total expected loss is only a fraction of the typical observed rate of loss of cloud nuclei in such a container (5 percent per hour), it is clear that the unrealistic assumption that the air in the container remains still must be abandoned.

### EFFECT OF CONVECTION CURRENTS ON PARTICLE LOSS

The loss of particles from air which is convectively circulating in a container obviously presents a rather intractable problem. However, an order-of-magnitude theory would be sufficient to indicate how such losses might depend on various factors.

#### A. The Convective Circulation

Consider a container with linear dimensions  $L$ ; and suppose that the temperature of one wall is  $\delta T^\circ\text{C}$  higher than the opposite one. An upward motion will occur near the warmer wall, a downward one near the colder. The momentum of the moving air is generated by



buoyancy forces in those regions where the temperature of the air has been significantly influenced by the neighboring wall, and suffers attrition as a result of viscous stresses. The temperature anomaly relative to the mean of a circulating parcel is reversed twice per revolution by heat conduction to and from the wall; no other mechanism significantly influences the temperature of the parcel. On the other hand, the momentum anomaly of the parcel is reversed twice per revolution as a result of the action of pressure forces which change the descending current to a horizontal one, and then into an ascending current. The scalar gradient of velocity from the region of maximum velocity to the wall is essentially unchanged during this process, while the gradient of temperature is reversed. Thus, although the diffusivity of momentum in air (the kinematic viscosity) is less than the diffusivity of heat, the non-steady aspect of the diffusion processes is of little significance for momentum, but is of the essence of the problem in the case of heat.

The vertical velocity will have an S-shaped distribution, vanishing in the middle and at the walls. Let  $v$  be the maximum velocity, which will occur somewhere between a wall and the center. Let the distance from the wall to this point be  $y$ . Then, air near the velocity maximum will traverse the length of the wall in time  $T = L/v$ . It is plausible that at the location where  $v$  occurs, the air temperature will be largely controlled by the wall. Otherwise, the convection current would not reach its maximum velocity there.

When a semi-infinite slab of material with a thermal diffusivity of  $\kappa$  and an initial uniform temperature  $T_1$  is placed in good thermal contact with a surface which is held at a fixed temperature  $T_0$ , heat diffuses through the material in a manner defined by

$$(T - T_0) = (T_1 - T_0) \operatorname{erf} (y / 2 \sqrt{\kappa t})$$

where  $y$  is the distance from the surface which is held at the fixed temperature  $T_0$ . The temperature at distance  $y$  asymptotes to its final value ( $T_0$ ), though not in an exponential manner. A reduction

$|T - T_0|$  by a factor of  $e^{-n}$  requires a period of  $(y^2/4\kappa q_n^2)$  seconds, where  $q_n$  is defined by:

$$\text{erf}(q_n) = e^{-n} \quad (q_1 = 0.339, q_2 = 0.120 \text{ etc.})$$

It will be assumed here that during its transit along the wall the air at position  $y$  spends a period of  $y^2/4\kappa q_1^2$  seconds, so that its temperature anomaly relative to the wall will be reduced by a factor of about  $e$ . Thus,

$$T - \frac{L}{v} = \frac{y^2}{4\kappa q_1^2} \cdot$$

or,

$$y^2 = \frac{4\kappa q_1^2}{v} \frac{L}{v}$$

Consider now the slab of air between the point  $y$  and the wall. Assume that its temperature is  $(\delta T/2)^\circ\text{C}$  warmer (or cooler) than the average of the entire volume. The buoyancy force acting on a column of length  $L$ , of unit breadth is then  $-gyL\delta\rho = 1/2gyL\rho \delta T/T$ . The only other force is the viscous stress at the wall, since at the location of the velocity maximum, there is no shear. Taking the gradient of velocity at the wall, as would be the case in a two-dimensional Poiseuille flow, the viscous stress along the wall, per unit breadth, is  $2\eta v L/y$ .

Hence,

$$\frac{1}{2} gyL\rho \frac{\delta T}{T} = \frac{2\eta v L}{y} \cdot$$

so that

$$v = \frac{gy^2}{4v} \frac{\delta T}{T}$$

where  $\nu$  is the kinematic viscosity of air. Substituting the earlier expression for  $y^2$ ,

$$\nu^2 = \frac{g\kappa q_1^2 L}{\nu} \frac{\delta T}{T}$$

and

$$y^4 = \frac{16\nu\kappa q_1^2 L}{g} \frac{T}{\delta T}$$

Taking  $g = 10^3 \text{ cm sec}^{-2}$ ,  $\kappa = 0.22 \text{ cm}^2 \text{ sec}^{-1}$ ,  $\nu = 0.15 \text{ cm}^2 \text{ sec}^{-1}$ ,  $L = 50 \text{ cm}$ ,  $q_1^2 = 0.115$ ,  $T = 300^\circ \text{K}$ , then  $\nu^2 = 28\delta T$ , and  $y^4 = 0.9/\delta T$ . Thus, for  $\delta T$  in the range  $0.01$  to  $0.1^\circ \text{C}$ , as seems plausible under laboratory conditions, the estimate of  $\nu$  lies in the range  $1/2$  to  $1-1/2 \text{ cm sec}^{-1}$ , and that of  $y$  in the range  $3$  to  $2 \text{ cm}$ .

Experimental work by Prandtl (1949) would indicate similar velocities, in the range  $0.7$  to  $2 \text{ cm sec}^{-1}$  under these conditions.

#### B. Particle Loss by Brownian Diffusion

In considering the effect of convection currents on particle losses, it would appear that a perfectly steady and laminar circulation of air would have little effect on the rate of loss. The particle-depleted sheath of air lying close to the wall would remain there, effectively shielding the bulk of the gas from much further particle loss. Unsteadiness in the flow, however, is very likely to occur, and a simple way to model the loss of particles is to suppose that the particle-depleted air very close to the wall is replaced by unmodified air every  $T'$  seconds. If this is so, the particles out to a distance  $x$  will be largely removed, if  $T' = x^2/4Dq_1^2$ , where  $D$  is the diffusivity of the particles; thus,  $x^2 = 4q_1^2 DT'$ . It will be assumed that  $T'$  is equal to  $mL/v_x$  where  $v_x$  is the convective velocity at distance  $x$  from the wall. This is equivalent to supposing that the sheet of air of depth  $x$  clings to a wall until it has traversed a distance equal to  $m$  times  $L$ , a typical linear dimension of the container. The number  $m$  may be expected to be of order unity.

The velocity at distance  $x$ ,  $v_x$  is about  $2vx/y$ , since  $x$  is quite small compared with  $y$ . Thus,

$$T' = mL y / 2vx$$

Combining the two expressions for  $T'$

$$\frac{x^2}{4q_1^2 D} = \frac{mLy}{2xv},$$

or

$$x^3 = \frac{2mLq_1^2 Dy}{v}.$$

From the above,

$$\begin{aligned} \frac{x}{T'} &= \frac{4q_1^2 D}{x} \\ &= 4q_1^2 D \left( \frac{v}{2mLq_1^2 Dy} \right)^{1/3} \\ &= \frac{2^{5/3} q_1^{4/3} D^{2/3}}{m^{1/3} L^{1/3}} \left( \frac{v}{y} \right)^{1/3} \end{aligned}$$

But,

$$\frac{v^4}{y^4} = \frac{g^3 q_1^2 \kappa L}{16v^3} \left( \frac{\delta T}{T} \right)^3$$

Hence,

$$\frac{x}{T'} = \frac{2^{4/3} q_1^{3/2} D^{2/3} \kappa^{1/12} g^{1/4}}{m^{1/3} L^{1/4} v^{1/4}} \left( \frac{\delta T}{T} \right)^{1/4}$$

Substituting the same values as earlier, assuming  $D = 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$ , an average value for cloud nuclei, and setting  $m = 1$ , this yields a value for  $x/T'$  of about  $2 \times 10^{-4} (\delta T)^{1/4}$ . If the area of the walls is  $A \text{ cm}^2$ , and the volume  $V \text{ cm}^3$ , the proportional loss

of particles per second will be about  $2 \times 10^{-4} A (\delta T)^{1/4} / V$ . In a typical bag container,  $A \approx 2 \times 10^4$ ,  $V = 3 \times 10^5$ , so that the loss rate would be  $5 (\delta T)^{1/4}$  percent per hour. For  $\delta T$  in the range 0.01 to 0.1°C, this corresponds to a range of 2 to 3 percent per hour.

The result is fairly insensitive to the value chosen for  $m$ . The fair agreement with observation (a total loss rate of about 5 percent per hour) may be fortuitous, but it gives some confidence to the prediction that the main cause of particle depletion is diffusion to the walls in the presence of convection currents, and that this loss will vary as  $(g\delta T/L)^{1/4}$ .

## CONCLUSION

When an aerosol is stored in a typical Faraday-cage type Mylar bag container with linear dimensions of the order of 50 cm, the cloud nucleus concentration is typically observed to decrease at a rate of about 5 percent per hour. If air motions occurring in the container are ignored, this cannot be explained by the combined action of diffusion, coagulation and sedimentation.

A rough theory indicates that in ordinary laboratory conditions (with a temperature contrast of  $10^{-1}$  to  $10^{-2}$ °C across the container), convection currents flow at velocities of the order of one cm sec<sup>-1</sup>; when the flushing of the air close to the walls is modeled, diffusion appears as by far the most important depletion mechanism; the theory predicts losses of 2 to 3 percent per hours. Since in a typical aerosol, coagulation and sedimentation together appear to cause a loss of less than 1 percent per hour, it is reasonable to attribute the rest of the observed loss (ca. 4 percent) to diffusion in the presence of convection currents.

According to the theory, the diffusional loss varies directly as the fourth root of both the temperature contrast across the container, and of the acceleration due to gravity, and inversely as the fourth root of the linear dimensions of the container. In an orbiting vehicle where  $g$  will be reduced by a factor of  $10^5$  below its terrestrial value, the expected loss rate due to diffusion with the

usual temperature contrasts present would be about  $4 \times 10^{-5/4} = 0.2$  percent; that due to sedimentation would be negligible. Taking the loss due to coagulation as 0.3 percent, the total loss rate would be about 0.5 percent per hour. Some further reduction in the loss rate could be achieved by making the container more isothermal, but little could be gained by any practicable increase in size.

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## **A.2 CONDENSATION NUCLEATION APPROACH 2**

### **A.2.1 Introduction**

The atmospheric nuclei consist of solid insoluble materials, soluble matter, and a mixture of soluble and insoluble particles. The last two groups of nuclei, being hygroscopic, are preferred as centers of condensation. Their efficiency in nucleation is determined by both their size and chemical nature. In atmosphere, the insoluble particles may obtain a hygroscopic film, either by capturing small solution droplets or by acting as condensation sites for trace impurities.

We have reported some time ago (R. Nelson and N. Gokhale, "Concentration of giant particles below cloud bases", Proceedings of the First National Conference on Weather Modification, 1968) on the presence of a significant number of giant particles in the size range 10 to 100 microns radii to be present below cloud bases. It is possible that these particles may be responsible for the initial growth of a few large drizzle size drops to initiate the collision-coalescence process in clouds. We found that these giant particles had some spots on their surfaces which were hygroscopic, however, the whole surface of the particle was not hygroscopic. In saturated atmosphere these particles developed small droplets by condensation on their surfaces.

It would be interesting, therefore, to study the growth of such particles by condensation and their efficiencies in collecting other cloud droplets. Under zero-g condition turbulence could be introduced into the chamber as necessary.

### **A.2.2 Objective**

Determine the nucleation efficiencies and early growth properties of soluble, insoluble, hydrophobic and "mixed" nuclei. These experiments include a large range of nuclei types size (including giant particles) distributions, and relative humidities.

### **A.2.3 Scientific Justification**

Kohler's theory has been applied with some success to explain the role of condensation nuclei in forming cloud droplets. However, numerical calculations of the cloud forming processes predict more nondisperse cloud droplet

spectra than are commonly observed. This is largely controlled by the size distribution and composition of cloud nuclei.

One of the outstanding problems in cloud microphysics is to account for the growth of sufficient droplets to a radius ( $>25\mu\text{m}$ ) beyond which they can continue to grow rapidly by coalescence to precipitation size. The role of giant condensation nuclei in producing such a few larger droplets is of utmost importance.

#### **A.2.4 Applications**

It is well known that nucleation processes are the key to weather modification. A better understanding of nuclei properties and their roles in cloud formation will permit (i) the effective use of condensation nuclei to modify and dissipate warm fog, (ii) modification of warm clouds by seeding with condensation nuclei, (iii) release of giant particles to initiate the collision coalescence process in clouds, (iv) better precipitation forecasting on the basis of measurements of the particulate matter in the air, (v) to know the effective type of nuclei, their size and concentrations to be introduced in a warm cloud for modification.

#### **A.2.5 Terrestrial Laboratory Limitations**

The earth bound cloud physics laboratories faced a number of restrictions in their experiments due to the suspension systems, influences on nucleation processes by the supports, convection, temperature and humidity variations in the chamber, and fall out due to gravitational forces. Particles from submicron to giant particles have significant fall velocities and hence without gravitational forces present, their nucleation efficiencies and time dependence can be studied. These determinations become difficult under limitations of a terrestrial laboratory.

#### **A.2.6 Zero-Gravity Opportunities**

Terrestrial diffusion chambers are restricted to a depth of a few cms by thermodynamic considerations. Therefore, the experiments performed in such a chamber are seriously limited by fallout. The different rates of growth of nuclei to cloud droplets can adequately be studied in the zero-gravity cloud physics laboratory. The "poisoning" of cloud nuclei, the role



of giant nuclei in forming few larger size droplets, their interactions with smaller size droplets and their growth are such experiments for which zero-gravity conditions offer distinct advantages.

#### **A.2.7 Quantification**

It is very difficult to predict accurately in advance the progress and success of any scientific experiment. However, as discussed in the earlier sections, the suggested experiments would be best carried out in a zero-g environment.

#### **A.2.8 Approach**

The critical growth properties of soluble (e. g. NaCl,  $(\text{NH}_4)_2 \text{SO}_4$ ,  $\text{Ca Cl}_2$ ,  $\text{H}_2\text{SO}_4$ ), insoluble (e. g. dust and smoke particles, AgI, CuS) and hydrophobic (e. g. Teflon) particles will be studied with the help of a continuous-flow thermal diffusion chamber (CFD). The sizes of the particles would vary from  $100 \text{ \AA}$  to  $100\mu$  in diameter.

The diffusion chamber as developed by Squires is shown in Figure A-3. Significant dimensions are the separation of the horizontal plates  $P_1$  and  $P_2$  ( $h = 1.3 \text{ cm}$ ), their length along the stream ( $45 \text{ cm}$ ), and their breadth ( $b = 29 \text{ cm}$ ). The sample is injected through a manifold (slit width  $8 \text{ cm}$ ,  $0.08 \text{ mm}$  deep) with a preconditioned sheath flow, thus confining the sample within the central  $2 \text{ mm}$  constant-supersaturation region of the chamber. The aerosol sample volume flow is between  $0$  and  $1 \text{ cm}^3 \text{ sec}^{-1}$ . The droplets exit from the chamber into an optical counter (Royco 225 or equivalent).

The overall pattern of air flow is shown in Figure A-4. The pump associated with the particle counter drives a circulating stream of air some  $330 \text{ cm}^3 \text{ sec}^{-1}$ , most of which forms an almost particle-free sheath surrounding the droplet-carrying stream from the diffusion chamber. A stream of  $42 \text{ cm}^3 \text{ sec}^{-1}$  is vented to the atmosphere through an orifice (B), the pressure at B being controlled by a flow resistor downstream. This flow is partly replaced by the metered bypass inflow (A), which consists of room air. The remainder of the  $42 \text{ cm}^3 \text{ sec}^{-1}$  required to replace the air exhausted at B passes through the diffusion cloud chamber; it consists mostly of the main flow (F), a metered flow of filtered room air, with some sample flow  $f$ . The pump

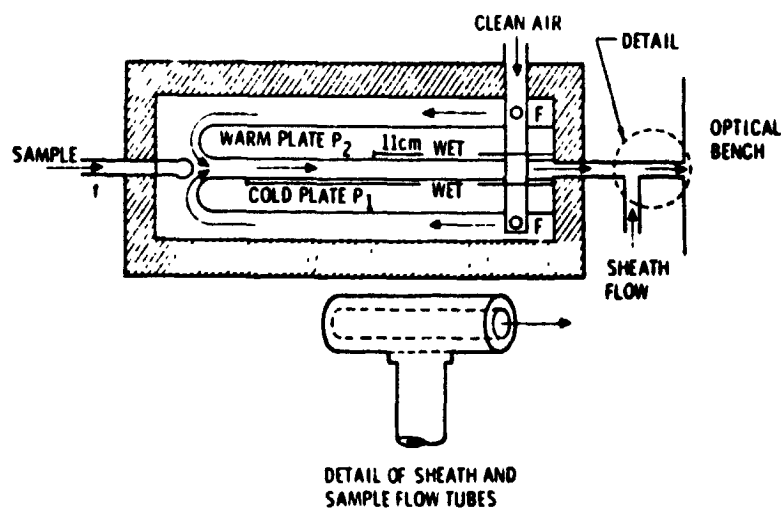


Figure A-3. Continuous-Flow Diffusion Chamber

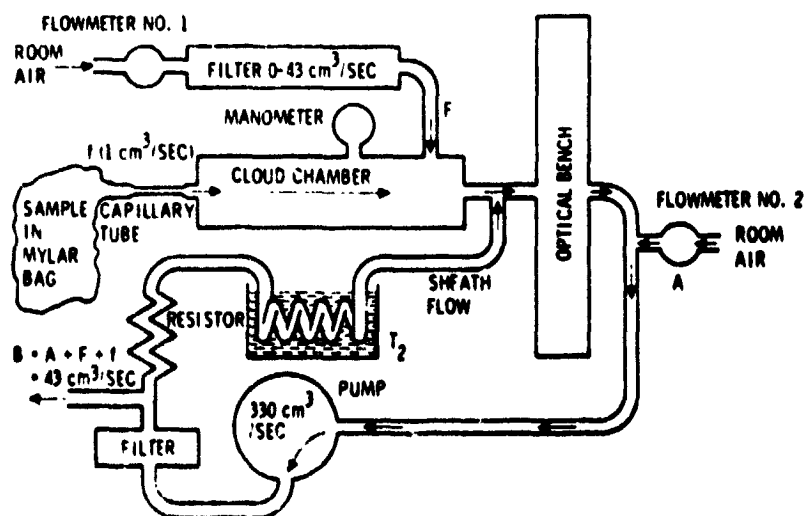


Figure A-4. Continuous-Flow Diffusion Chamber Air Flow

unavoidably heats the air. To avoid particle counterheating and possible droplet evaporation, the sheath flow air is cooled to the temperature of the top plate,  $T_2$ . The temperature difference between the two plates is measured to  $0.05^\circ\text{C}$ .

The chamber operates at a deficit pressure  $1.33 \times 10^3 \text{ N/m}^2$  (10 torr) relative to the air supply pressures. In an experiment where it is desired that the super-saturation be constant to within 0.02 percent, the maximum rate of change of pressure which can be tolerated in a chamber 1 cm deep (1/2 sec time constant) is around  $50 \text{ N/m}^2 \text{ sec}$  (0.38 torr/sec) relative to a total pressure of  $8 \times 10^4 \text{ N/m}^2$  (610 torr). For the zero-gravity cloud physics laboratory, the main air flow would be derived from stored air compressed in a cylinder (flow rate up to  $4 \times 10^{-5} \text{ m}^3/\text{sec}$  (cfh) at one standard atmosphere) and all exhausted air would be delivered to a sump tank.

A mylar bag is filled to its 2/3 capacity with high purity air passed through a desiccator and absolute filter. The nuclei will then be introduced into the bag. Under different coagulation times, different size particles will be available for testing in the conditioning chamber. The nucleation time of several seconds to several minutes will be required for nuclei activation and growth, depending on their sizes and the supersaturation in the chamber.

To simulate condition in a cloud, nuclei must be tested for their efficiency at small supersaturations. Hence, the continuous flow diffusion chamber is especially suited for this investigation. The supersaturation profile between the plates can be calculated from the upper and lower plate temperatures and chamber pressure. Optical particle counters will be used to determine the nuclei sizes and numbers.

#### **A.2.9 Experiment Parameters**

The important parameters along with the desired variations for each parameter are given below. This class of experiments concerning condensation nuclei can be divided into five basic groups: (1) soluble nuclei, (2) insoluble nuclei, (3) hydrophobic nuclei, (4) "mixed nuclei," and (5) "giant" nuclei.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	
Type	6
Pollutant	4
Pressure	3
Temperature	4
Relative humidity	4
Charge	
Rate of cooling	
Time	6
Sound	
Electric field	
Nuclear radiation	
Adsorption	4
Turbulence	*
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	3
Surface tension	
Aerosol age	3
History	3
Ion level	
Initial conditions	
Kinetic energy	
Cases	
Spin rate	

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\*Mentioned in context but not specified.

#### **A.2.10 Procedure**

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with the correlation of this to information to the low-g environment. Additional effort will be required to make these timelines operational, effective and efficient. (See Figure A-5.)

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<u>Preparation</u>	
● Set up experiment	30
● Control program	15
● Check out equipment	15
<u>Operations</u>	
● Purge cloud chamber	10
● Aerosol generation	7
● Establish temperature	} 10
● Establish flow	
● Calibrations	7
● Obtain data (observations)	50
● Data dump	1
<u>Analysis and Cleanup</u>	
● Data evaluation	40
● Cleanup and shutdown	20

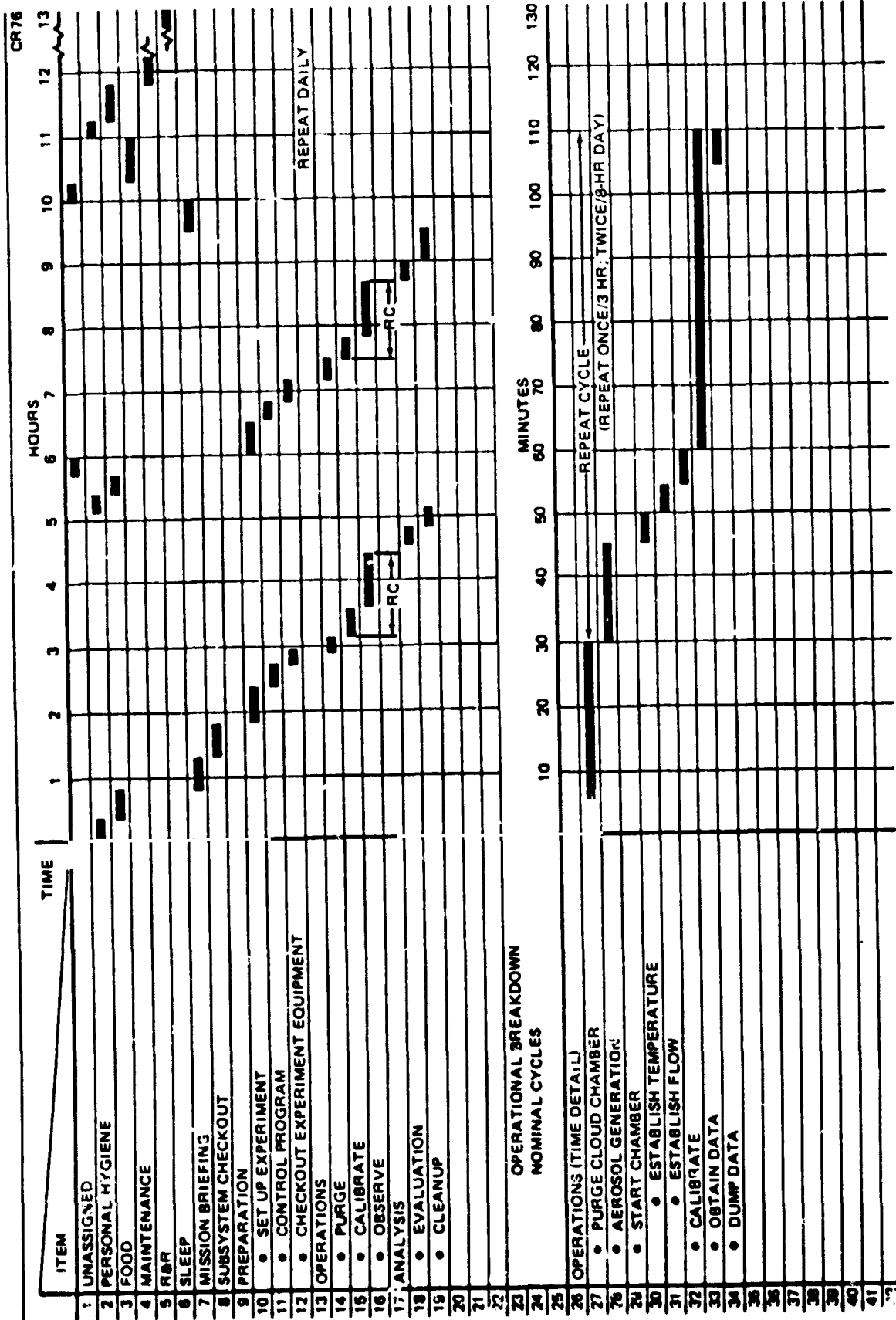


Figure A-5. Activity Timeline (One Day) Experiment Class 1 Condensation Nuclei

### A.3 CONDENSATION NUCLEATION APPROACH 3

#### A.3.1 Introduction

The ease with which a condensation nucleus forms a water droplet is dependent on a variety of factors, including solubility, hygroscopicity, size and, for insoluble particles, the wettability or contact angle of a water drop on the material. Several theoretical treatments have been proposed to describe the activation spectra of natural condensation nuclei. One model of nucleation, the Volmer theory (1939), predicts a dependence of critical supersaturation\* on the contact angle that the liquid makes with the substrate. Thus, a particle exhibiting hydrophobic properties would not be expected to participate in most cloud systems. In fact, materials which have a contact angle greater than  $6^\circ$  are often considered noneffective as condensation nuclei in clouds and fog (McDonald, 1964).

Recent studies have indicated, however, that such nominally hydrophobic materials as Teflon, wax and paraffin (see, e. g., Jiusto and Kocmond, 1968) can effectively behave as condensation nuclei at relatively low supersaturations. The view is that adsorption of water vapor on the surface of the nucleus (sometimes taking place over substantial periods of time) overcomes the hydrophobic characteristics of the particle. Under these conditions, condensation and droplet growth proceed as if the nucleus were a fully wettable substance.

At the other extreme of particle activity, one must view the importance of soluble hygroscopic nuclei in natural cloud systems. Kohlers (1926) theory of heterogeneous nucleation of droplets on soluble nuclei basically correlates nucleus size with critical supersaturation; for hygroscopic nuclei, such as NaCl, the theory is quite straightforward and widely accepted for modeling studies involving droplet growth in clouds.

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\*The supersaturation beyond which droplet growth proceeds without limit.



Investigations of particle activity are scarce primarily because of difficulties in developing suitable instrumentation, obtaining accurate particle size distributions, and in obtaining chemically pure materials for testing. Experiments performed at the Second International Workshop on Condensation and Ice Nuclei (IWCIN), Ft. Collins, Colorado, August 1970, showed some rather large discrepancies between the predicted and experimentally determined nucleating efficiencies of NaCl nuclei. The results suggest that a NaCl nucleus must be about twice as large as predicted from theory in order to act as a cloud nucleus at the desired supersaturation. Additional experiments performed under more carefully controlled laboratory conditions at the University of Minnesota pointed to even greater differences (Katz and Kocmond, 1973).

It is probable that at least a part of the discrepancy between theory and experiment can be ascribed to surface impurities which modify the properties of the nuclei being tested. Producing chemically pure nuclei that are void of any surface contaminants is indeed a challenging problem. Once the particles are formed, examining their nucleating behavior over sufficient periods of time in a cloud chamber has often been impossible in the terrestrial laboratory.

In the proposed Zero-G experiments, droplet growth in the supersaturated environment can be studied for periods of time that are comparable to those found in natural clouds. Especially for hygroscopic nuclei, particle activity can be examined at supersaturations of a few hundredths of a percent, a value that is widely accepted as being representative of natural stratus clouds and fog.

#### A.3.2 Objective

The basic objective of the experiments will be to examine the nucleation efficiencies of soluble, insoluble and hydrophobic nuclei. Experimental results obtained with the continuous flow diffusion chamber (CFD) will be compared with the theoretically predicted values of particle activity. Since basic chamber systems will be interchangeable, a static diffusion chamber (SDC) can be available for alternate use. An equally important objective of these experiments will be to observe the activation spectra of nuclei at supersaturations typical of stratus clouds and fog (i. e. ,  $<0.1$  percent S).

#### A.3.3 Scientific Justification

Data obtained from the experiments will contribute to our understanding of the nucleation properties of several important classes of nuclei (i. e., soluble, insoluble and hydrophobic particles). For the first time, droplet growth can be observed over extended periods of time and at extremely low supersaturations (i. e.,  $<0.1$  percent S). This information will be helpful in understanding the early droplet growth processes in clouds and will greatly benefit the researcher in establishing the nucleating behavior of common seeding agents used in weather modification. Questions regarding differences between nucleation behavior in experiment and in theory may partially be resolved by this critical series of tests. Finally, better predictions of cloud and precipitation behavior in the natural atmosphere may be possible as a result of data obtained in these tests.

#### A.3.4 Applications

The interaction of nuclei with water vapor in the atmosphere forms the basis for all precipitation processes. Knowledge gained from these experiments will provide needed comparisons between experimentally derived and theoretically predicted values of particle activity. This information will be essential to the researcher and applied meteorologist desiring to predict the effect of seeding agents on warm clouds and fog. Additional benefits will be derived by those attempting to predict the impact of certain pollutants on cloud and fog formation, persistence and dissipation. Questions relating to inadvertent weather modification due to anthropogenic sources of nuclei can more realistically be answered with improved knowledge of the nucleating efficiencies of various types of nuclei.

#### A.3.5 Terrestrial Laboratory Limitations

The limitations imposed on terrestrial diffusion chambers are well known. Gravitational forces result in the fairly rapid fallout of growing droplets within the chamber and so performance is limited. At low supersaturations ( $<0.1$  percent S) nucleation rates are quite slow and frequently particles are lost from the viewing volume before they have grown to observable sizes. At slightly higher supersaturations, many of the larger and more hygroscopic nuclei grow and settle out of the sensitive volume before the less active nuclei

reach detectable sizes. It is not uncommon in the terrestrial laboratory to obtain an apparent nucleus count that is substantially lower than the true value.

#### A.3.6 Zero-Gravity Opportunities

Zero-gravity conditions are particularly well suited for observing nucleation behavior and droplet growth over extended lengths of time. Since fallout is much less of a problem, observations can be made at low supersaturations and on slow-to-activate hydrophobic nuclei. Gravity conditions in the terrestrial laboratory using the static or, in some cases, the continuous flow diffusion chamber do not permit such experiments to be performed.

#### A.3.7 Quantification

Problems involving nucleation theory continue to be among the most pressing to cloud physicists and researchers in the atmospheric sciences. The unique opportunities offered by the Zero-G environment cannot be overemphasized. By removing the constraints which limit the utility of diffusion chambers in terrestrial laboratories essential information can be obtained on nucleation behavior and initial growth rates of particles at low supersaturations. Comparisons of the data with nucleation theory will be of special relevance to the entire scientific community. The prospects of achieving this very substantial goal warrant our most aggressive efforts to get the experiments underway. An additional important feature of the Zero-G environment is that the data can be acquired with equipment that is available today. Several years of development time and substantial costs would be involved in developing a suitable chamber for comparable measurements in the terrestrial laboratory.

#### A.3.8 Approach

##### A.3.8.1 General

The basic approach will be to produce nuclei of the desired type and size and to introduce these nuclei into a continuous flow diffusion chamber (CFD). An optical sensor will also be used to provide information on droplet sizes achieved within the CFD. Measurements of nucleation efficiency will be compared with theoretical data and will be performed over a broad range of chamber conditions; particular attention will be given to operating at low supersaturations (i. e.,  $< 0.1$  percent) typically found in stratus clouds and fog.

A static diffusion chamber utilizing an automated data recording system could be utilized. The problems associated with diffusion chamber design have been discussed by several workers in the field, including Twomey (1959), Saxena (1970), Fitzgerald (1970) and Squires (1971). Basically these discussions warn the experimenter that care must be taken to avoid transient high supersaturations (through contact of the sample air with warm moist or cool dry surfaces), that the chamber depth should not exceed 1 cm, that excessive nucleus concentrations ( $>10^3 \text{ cm}^{-3}$ ) can lead to erroneous results, and that the maximum usable supersaturation of a static chamber in the terrestrial laboratory is about 0.2 percent S. Although the Zero-G environment eliminates the last consideration and greatly enhances the overall utility of the static chamber, the recent identification and further development of the continuous flow system (Squires, NASA CR-129013) makes the CFD especially well suited to this class of experiments. As part of supporting studies by Desert Research Institute ample documentation has been provided on the design and operating principle of the CFD (NASA CR-129013) and so additional discussion of this instrument system will not be presented here. It will be assumed in the description which follows that the CFD system will be used for this class of experiments and that the SDL will be available on a standby basis.

As noted earlier, previous investigations aimed specifically at understanding the nucleation efficiencies of soluble, insoluble and hydrophobic nuclei are scarce. Attempts to verify nucleation theory by conducting carefully controlled experiments in the laboratory are even less plentiful. In the Introduction, some past efforts along these lines are described. From a theoretical standpoint, the Kohler theory (1926) has successfully been applied to describe droplet growth on soluble hygroscopic nuclei. For insoluble particles, the Volmer theory (1939) predicts a dependence of particle behavior on the contact angle that water makes with the substrate. Thus, hydrophobic particles which have a large contact angle are not expected to participate in natural cloud and fog formation.

Past experimental investigations by Fletcher (1959) using AgI nuclei showed an activation supersaturation of 2.5 percent, in agreement with contact angle theory. These experiments, however, employed the use of an expansion chamber which could not produce supersaturated conditions over long periods

of time. Twomey and Severence (1964) and Twomey (1965) reported experiments where natural aerosol sizes were estimated using a diffusion battery and related to critical supersaturations. Interpretation of these results is rather difficult, however, since the nuclei sampled were of a mixed nature and their composition was unknown. Measurements of the activation supersaturation of silver iodide and wax nuclei were made by Juisto and Kocmond (1968) using a static diffusion chamber. The results obtained indicated supersaturation values considerably lower than those predicted by the Volmer theory.

Later, at the Second International Workshop on Condensation and Ice Nuclei (IWICN) held in Ft. Collins, Colorado, August 1970, several experiments were run with thermal diffusion chambers in which artificially generated nuclei of nominally uniform composition were introduced into a large 54 m<sup>3</sup> holding system and distributed to the instruments through 20 mm copper ductwork. Sodium chloride, silver iodide, and Teflon aerosols were produced for the experiments and their size distributions measured with the Minnesota Aerosol Analyzing System (MAAS) (Whitby and Husar, 1971a). In these tests significant differences were found between experimentally observed and theoretically derived nucleating efficiencies for NaCl nuclei. The experimental values of nucleus size required for activation to droplet growth were found to be twice as great as theory would predict. Even larger differences were found when some of the experiments were repeated during a joint workshop at the University of Minnesota (Katz and Kocmond, 1973). In these tests nuclei were produced from reagent grade NaCl either by heating or from a pneumatic nebulizer. After preparation, the nuclei were stored in a 4.5 m<sup>3</sup> nylon bag. Part of the nuclei sample was then transferred to a thermal diffusion chamber, a Gardner Small Particle Detector, and also to a Whitby Aerosol Analyzer (WAA). Two units of the WAA were on hand to provide additional comparisons. From a combination of these data, the relationship was derived between supersaturation and necessary minimum size of the particles for nucleation. The experimental values indicated that at a given supersaturation a NaCl particle has to be two to three times larger than theory predicts in order to be active as a cloud condensation nucleus.

It is probable that in these tests some or most of the observed differences between theory and experiment can be ascribed to surface impurities on the nuclei which in turn cause differences in the activation spectra. Another source of error is the size analysis of the nuclei being observed. In spite of these differences, the discrepancies between experiment and theory are quite large and need to be resolved if our understanding of nucleation theory is to improve. The Zero-G environment provides this opportunity as long as chemically pure materials can be prepared and accurate size distributions can be obtained.

For the proposed experiments involving nucleation of soluble, insoluble, and hydrophobic nuclei, some advanced development work is needed to derive methods for particle preparation and dispersion. (It is assumed that additional testing and development of the CFD will be accomplished in terrestrial laboratories prior to use in zero-gravity experiments. This will include proper thermal control, water supply delivery, acceptable weight and power demand and adequate reliability of all chamber operations.) Methods of preparing nuclei (large and giant as well as Aitken) that are free of all surface contamination must be devised. Exceptionally high purity materials and special handling techniques must still be developed in order to assure meaningful comparisons of experiment with theory. In particular, teflon powders that are free of catalytic impurities will be required for the hydrophobic nucleus tests (pyrolysis of teflon is not recommended since this procedure modifies the surface properties of the nucleus).

Once the materials and powders are developed, the problems of dispersion can be tested further; however, these problems appear less formidable. Additional experiments must be conducted in the laboratory to specify vaporization temperatures and coagulation times for various NaCl size distributions; solution concentrations for insoluble nuclei such as dioctyl phthalate; and aeration procedures for dispersing the teflon powder.

#### A. 3. 8. 2 Experiment Parameters

The important parameters along with the desired variations for each parameter are given below. This class of experiments concerning condensation nuclei can be divided into three basic groups: 1) soluble nuclei (NaCl), 2) insoluble nuclei (DOP, AgI) and 3) hydrophobic nuclei (Teflon).

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	
Type	5
Pollutant	3
Pressure	2
Temperature	3
Relative humidity	4
Charge	
Rate of cooling	
Time	6
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	*
History	*
Ion level	
Initial conditions	
Kinetic energy	
Gases	2
Spin rate	

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\*Listed but not specified.

#### A.3.9 Procedure

In a typical experiment, high purity air or nitrogen will be passed through a desiccator, absolute filter, and into the conditioning chamber. The conditioning chamber may be a metalized mylar bag or other holding device. The size of the bag varies, however, in a terrestrial laboratory a bag of 5 to 10 m<sup>3</sup> is often used to minimize aerosol losses during an experiment. Once the bag is partially filled with particle free air, nuclei of the desired chemical composition are generated and introduced into the bag. The amount of time that the aerosol is allowed to reside in coagulation tubes will partly determine the size distribution of the particulates; long residence times will result in nuclei of up to a few tenths of a micron in diameter while very short coagulation times will produce nuclei in the hundredths of a micron size range. In the case of NaCl aerosols, where electrical heating can be used, the temperature of vaporization will also influence the result. The exact relationships of flow rates, vaporization temperature, and coagulation times vary depending on the conditions desired for a test. The nuclei may be further modified by passing them through pollutant gases, an irradiation source, evaporation retardants or diffusion battery to remove very small particles.

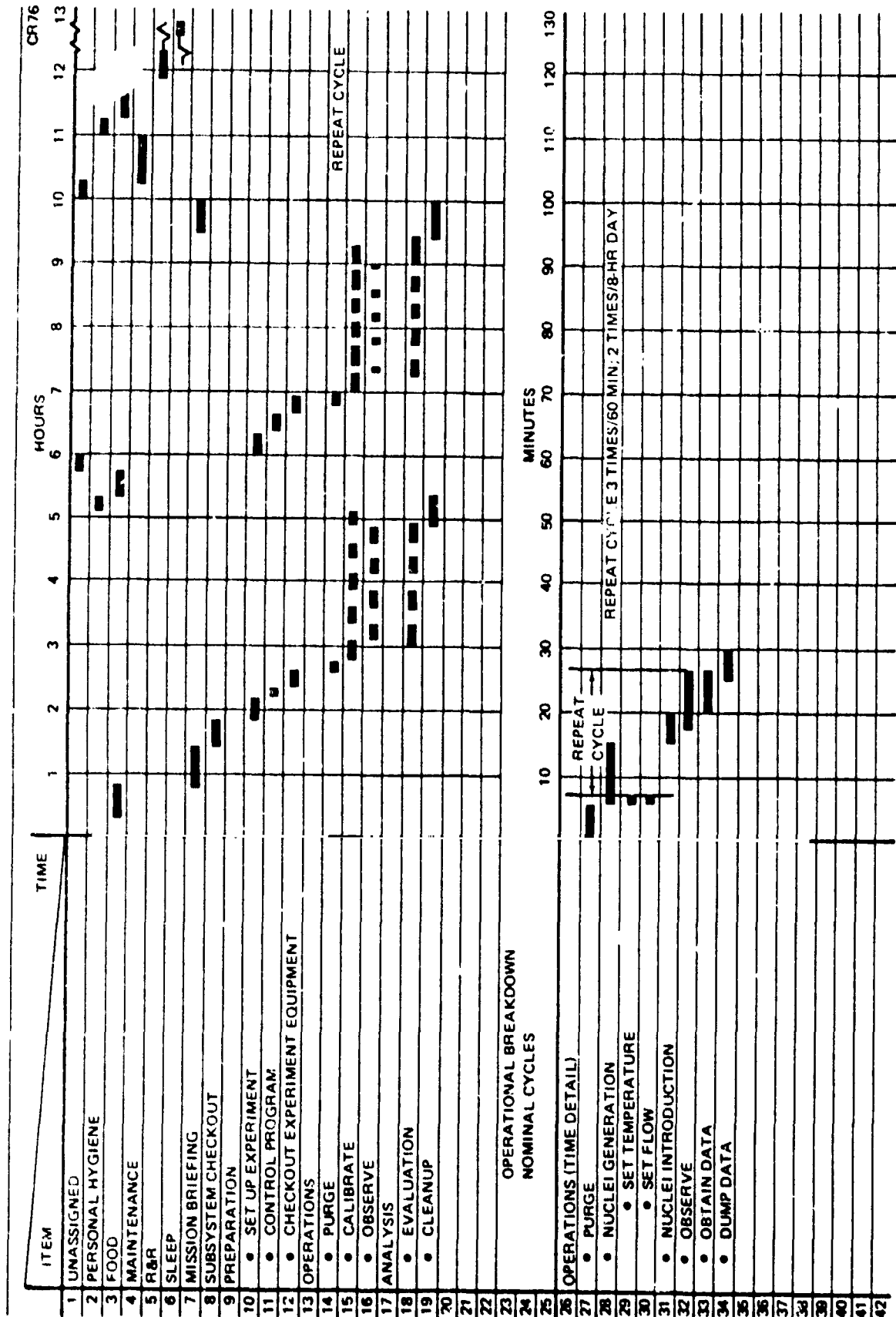
After nuclei of the desired type and size are introduced into the conditioning chamber, samples are passed into an aerosol analyzer (EAA) and at the same time admitted into the thermal diffusion chamber and total nucleus counter. Depending on the aerosol type and the conditions of supersaturation in the chamber, nucleation may take place in several seconds or several minutes. For most atmospheric nuclei, droplet growth to detectable sizes occur in six or seven seconds. Following an observation, the aerosol is flushed into a holding system for latter disposal.

Tabulation of general activity details are given below followed by a representative activity timeline. The event sequence and indicated times are based on knowledge of terrestrial requirements and restrictions with thought to translating this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient. (See Figure A-6.)



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● <u>Set up equipment and check out all systems</u>  This will include setting up the CFD or SDC chambers (depending on the instrument desired), testing for proper flow through regulators and flowmeters, preparing aerosol generator or nucleus sample for particle production, insuring that the optical sensor is functional and checking out data gathering systems.</li> <li>● Purge CFD (or SDC), conditioning chamber, and electrical aerosol analyzer with purified nitrogen or compressed air and check for zero.</li> <li>● Generate nuclei for experiment. Normally this will involve terminating all gas flow and producing nuclei of the desired type and size. Either NaCl, DOP, or Teflon aerosol<sup>o</sup> will be used in an experiment.</li> <li>● Set supersaturation. While nuclei are being prepared, set CFD to desired supersaturation for the test and set flow in chamber.</li> <li>● Introduce nuclei and obtain data on nucleation efficiency, droplet growth, and particle size distribution.  After producing nuclei and allowing them to reside in the coagulation tube and conditioning chamber, they are passed into the CFD and at the same time a portion of the sample is directed to the EAA (particle size analysis). The flow of the CFD will be adjusted to allow ample residence time of the nuclei within the chamber. After growing to droplet sizes (<math>&gt;2\mu\text{m}</math>), the sample will pass into an optical sensor for drop size and CNC measurements.</li> <li>● Following the determination of CNC, droplet sizes and particle distribution, the sample may be flushed from the system.</li> </ul>	<p>40</p> <p>5</p> <p>&lt;1</p> <p>5</p> <p>1</p>



### A.3.10 References

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Appendix B  
CLASS 2  
ICE NUCLEATION

B.1 ICE NUCLEATION APPROACH 1

B.1.1 Introduction

Among the population of atmospheric aerosol particles, ice forming nuclei occupy only a minute fraction. However, they are important because they form ice crystals in supercooled clouds and trigger a thermodynamically supported change, i. e., glaciation. This process frequently leads the cloud to develop precipitation. Ice-phase weather modification is based on this phenomenon. Whenever the natural ice-forming process is inefficient in supercooled clouds or cloud systems, introduction of artificial ice nuclei helps the thermodynamical change to initiate and results in modification in the cloud structure, energy balance, and often additional precipitation.

In order to understand the cloud process and to further modify it in a desirable direction, we must have exact knowledge of this process of ice nucleation.

The mechanism of ice formation on the nuclei is complex. The macroscopic modes of ice nucleation are of our direct concern when we are to apply our knowledge to the atmospheric processes, although the micromechanisms are indirectly connected. The mechanism of ice nucleation may be classified in terms of homogeneity of the initial phase or phases involved such as homogeneous or heterogeneous ice nucleation. Another way to categorize is through their histories. The formation of ice phase for the first time without any remains of past history may be called the primary ice nucleation, and if it is affected by the past history, it may be called the secondary ice nucleation. The former is the ice nucleation of the type most commonly discussed, and the latter refer to processes like ice formation with memory effect, fragmentation and splintering. This study concerns the heterogeneous ice nucleation of the primary kind.

The ice nucleation mechanism which attracted the largest attention during the past several years is probably freezing of supercooled cloud droplets upon contact with nucleus particles (contact nucleation; Gokhale and Gould, 1968; Gokhale and Spengler, 1972; Isaac and Douglas, 1972; Alkezweeny, 1971)

This nucleation mechanism was also observed on organics and distinguished from the nucleation of freezing by submerged particles (Fukuta et al, 1971; Fukuta, 1972). Ice crystals formed by the contact of nuclei with supercooled droplets apparently show a double-plate form, depending on the temperature (Weickmann, et al. 1970 Auer, 1972). Fletcher (1970) suggested contact nucleation as a possible variation of freezing nucleation by submerged nucleus particles. Of course, this does not explain the difference of the nucleation behavior observed between a nucleus particle colliding with a supercooled droplet and a nucleus particle sitting on the surface of the droplet (Fukuta, et al. 1971).

Two long-known mechanisms of ice nucleation, i. e., sublimation and freezing (including nucleation on the surface of a nucleus in cold air at above water saturation, which is sometimes referred to as sorption nucleation) have been subjects of experimental studies (Bryant et al, 1959; Edwards and Evans, 1960 and 1961; Fukuta, 1958; Layton and Steger, 1969; Ramachandra Murty and Ramana Murty, 1972; Vali, 1971) including size effect (Gerber, 1972) and of theoretical studies (Davis, 1972; Fletcher, 1958, 1969, 1970, 1972). The improved formulation of homogeneous nucleation theory has been of value to these studies (Abraham, 1968, Kiang et al, 1971).

Furthermore, the ice nucleabilities of ice nucleus compounds depend also on the particle size, the method of preparation, and the environment in which the compounds have been placed. The ice nucleation of the kind in question here, therefore, is the result of combined basic mechanisms under the particle and environmental conditions. In view of the fact that all of these factors are of a highly delicate nature, the current position of the ice nucleation field is rather chaotic; new studies have been bringing more confusion rather than clarification.

Under the conditions of terrestrial laboratories, a time-dependent factor of ice nucleation at a fixed space coordinate becomes also space-dependent by the time the detection method is applied due to the fact that ice crystals as the result of the nucleation are subjected to the gravitational field to fall and spread. Efforts of simplifying the space-time dependence in the terrestrial laboratories frequently lead to sacrifice of one of the factors.

What is most needed in this field is clear cut conditions of nuclei activation, particularly in terms of supersaturation and fog droplet factors, since nuclei history including the preparation can be controlled reasonably well. The Zero-Gravity Cloud Physics Laboratory provides a unique opportunity of testing ice nucleation under clearly defined conditions without sacrificing some of the factors commonly observed in terrestrial laboratories.

#### **B. 1.2 Objective**

The objective of this study is to determine the relative importance of contact, condensation freezing, and sublimation nucleation of ice. Absolute nucleation efficiencies will also be examined as a function of the nuclei type, size, and cloud condensation history. During the investigation, the time lag phenomenon of ice nucleation will be checked.

#### **B. 1.3 Scientific Justification**

Ice nucleation is the start of ice phase changes in clouds. All the phenomena directly or indirectly related to the changes, therefore, will be affected by the result of the nucleation. Ice nucleation in clouds is complex, having a number of mechanisms simultaneously in action. The desirable approach to the phenomenon of such a complexity is to analyze it with respect to each individual mechanism and to assemble all the contributions from different mechanisms back to the original overall ice nucleation phenomenon, taking differences of cloud conditions into account. This requires quantitative data of all the nucleation mechanisms involved, such as the contact, condensation freezing, and sublimation nucleation determined under clear-cut conditions. The unique Zero Gravity Laboratory condition permits such a study.

As recent weather modification studies employ cloud models, incorporation of accurate data improves the predicting power of the model and allows one to find the particular seeding method which leads to the optimum result under the given cloud condition.

#### B. 1.4 Applications

These data are important in efforts to predict and to modify weather for all ice phase precipitation processes such as occur, for example, in snow, hail, and cold fogs. Proper seeding decisions will permit the redistribution and augmentation of snow, the clearing of supercooled fogs in airports, the minimizing of hail damage, and the reduction of the power of the destructive winds of hurricanes. These experimental data will contribute to the "what" (type of seeding material), "when" (in the precipitation cycle), "how much" (seeding material) and "where" (to seed) decisions involved in weather modification.

#### B. 1.5 Terrestrial Laboratory Limitations

The ice nucleation tests depend on the ice crystals formed. When ice nucleation takes place in cloud chambers on the ground, ice crystals formed move away from the points of nucleation due to the gravitational settling. Therefore, even if the environmental conditions such as supersaturation and temperature are only a function of the space coordinate and independent of time, it is extremely difficult to determine the original conditions of ice nucleation from the observed ice crystals. Using a supporting surface like the hydrophobed glass adds complications. The ice crystal falling in cloud chambers makes the analysis of nucleation modes difficult in the terrestrial laboratories.

#### B. 1.6 Zero-Gravity Opportunities

For this study, the low-gravity condition in a space laboratory helps the nucleated ice crystals stay in their original positions and presents an opportunity to perform accurate experiments. There is no need to say that a study of this kind depends solely on its accuracy, since the questions to be answered are "to what extent"? or "how many percent"?

### B.1.7 Approach

There are two basic types of experiments to be carried out. First is the experiment in which the number of ice nucleation for each of the three possible mechanisms will be determined with respect to the temperature, supersaturation and fog droplet condition such as size and number concentration, having the temperature fixed. This will clarify basic mechanisms involved in ice nucleation of each compound. The second type is to simulate the ice nucleation behavior during the cloud processes involving cloud condensation before ice nucleation, having the temperature (cooling) this time as the variable and the supersaturation with respect to water fixed (the condition where cloud droplets continue to grow). In the experiment of the second type, ice nuclei introduction into a particular part of the cloud cycle will be simulated.

Ice nuclei samples are selected organic and inorganic compounds, and clay minerals including actual soils.

For this study, a special thermal diffusion chamber will have to be built and be used at sub-freezing temperatures without flow inside (see Figure B-1). The chamber is square in shape (25 cm x 25 cm) with one end connected by a

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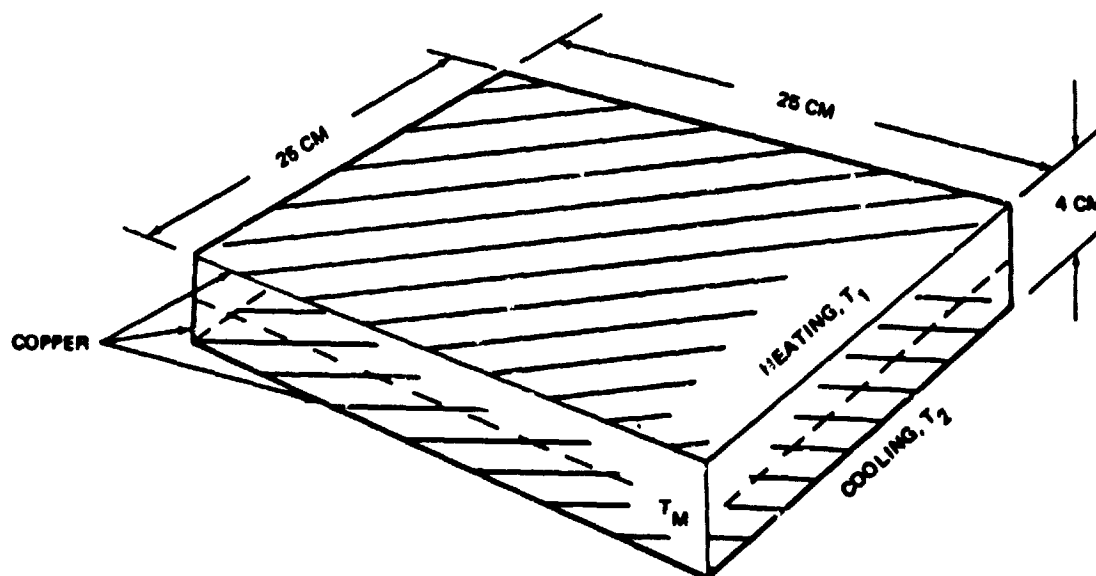


Figure B-1. Sketch of University of Denver Static Thermal Diffusion Chamber



thermal conductor wall (4 cm high). Three other sides are made of transparent thermal insulators. The top edge opposite to the side conductor wall will be kept warmest ( $T_1$ ) in the chamber whereas the corresponding bottom edge will be maintained as coldest ( $T_2$ ). There will be thermal gradients along the top and bottom plates normal to the side conductor wall as well as in the direction normal to both plates. The median plane is devoid of thermal gradient (the temperature is  $T_M$ ) but maintains varying supersaturation with respect to ice along the plane and in the direction perpendicular to the side conductor wall.  $T_1$  and  $T_2$  must be adjustable to any level of the range in question.

The diffusion chamber for this study is basically the same as that for cloud condensation nuclei studies. The main difference, except the additional thermal gradients, is that ice sheets will be used instead of liquid water at the inner surfaces of top and bottom plates. The ice sheets are frozen water in filter papers. They have to be renewed every time by supplying water with a wick after warming.

The sample smoke will stay still in the chamber without flowing.

The experimental procedure is as follows. Smoke particles generated by means of the modified LaMer-Sinclair Generator (Class 13, Instrumentation) will be kept in the smoke box. Soil samples must be prepared in the terrestrial laboratories by dispersing them in air and gathering the slow settling fraction (fine particles) in plastic bottles. Clay minerals must be ground and put in a plastic bottle while they are on the ground. The plastic container has a coarse filter and a tube. For smoke nuclei generation, the bottle will be shaken vigorously and then squeezed. The filter will retain large particles in the bottle.

Two types of experiments will be performed.

The first type of experiment is as follows. The Denver University Static Thermal Diffusion Chamber (DU-SDI) will be cooled to the temperature of study having no temperature difference between  $T_1$  and  $T_2$ . A small volume of the sample smoke dispersed in very dry air to avoid the transient supersaturation effect will be introduced into the DU-SDI which is devoid of cloud condensation nuclei (CCN). The smoke particles will spread and become quiescent as the eddies die down. If the mixing between the smoke and the chamber air does not occur properly, an additional stirring will be provided. Then,  $T_1$  will be slowly raised having  $T_2$  simultaneously cooled. During this process,  $T_M$  along the median plane will remain unchanged, but there develops a supersaturation profile along the plane. The ice crystals nucleated in the plane will be illuminated with a laser beam of a known cross section and be photographed at a set time interval. The number concentration of ice crystals nucleated will be estimated from the number in a unit length of the beam on the photograph. The supersaturation profile can be estimated from the temperature distribution on the top and bottom plates. This will continue until a time considerably after the point of fog droplet appearance. The increment in the number of ice crystals after the saturation point with respect to water can be attributed to the condensation freezing mechanism, whereas the number before the dew point is due to the sublimation ice nucleation.

An identical run will be made with a known number concentration of CCN and the difference in the number of ice crystals formed between these identical runs with and without CCN in the zone supersaturated with respect to water can be attributed to the contact mechanism of ice nucleation. These experiments will be repeated at different levels of  $T_M$ .

The second type experiment is to examine ice nucleation during a simulated process of cloud formation. It is also possible to study the nucleation during the simulated process of cloud dissipation. The difference in number of ice nucleations between the formation and dissipation processes of clouds may provide a clue to the diffusiophoretic and thermophoretic contributions to the contact ice nucleation. However, this study only concerns the formation process at this stage. The experimental procedure is as follows. A small

volume of dry smoke sample in the smoke box will be introduced into DU-SDI which is kept at the temperature of the cloud base. The chamber is also free of any thermal gradient and contains room air. After having the sample smoke uniformly dispersed in the chamber,  $T_1$  and  $T_2$  will be lowered at different rates ( $T_1$  lowers faster than  $T_2$ ) so that the supersaturation profile develops along the median plane while  $T_M$  continues to lower. When the temperatures of both plates reach  $0^\circ\text{C}$ , the water in the filter papers inside of the plates freezes with the help of attached guides which are capillary waters chilled strongly at the other end to maintain the ice phase. When the chamber wall temperatures become lower than  $0^\circ\text{C}$ , the stored ices grow and freeze the water in the filter paper. The cooling modes of both plates will change so as to maintain the continuity of the supersaturation with respect to water. The ice crystals formed on the median plane will be photographed by illuminating with the laser beam at a set time interval. This process will continue until the temperature becomes so low that ice nucleation is no longer important there. Variables are sample type, cooling rate, supersaturation, and composition and concentration of CCN. The suggested ranges of variables are:

$T_1, T_2$ : -35 and  $25^\circ\text{C}$

$T_M$ : -25 and  $25^\circ\text{C}$

Samples: 1, 5-Dihydroxynaphthalene and Phloroglucinol for organics

AgI for inorganics

Kaolinite for clay minerals

and one or two soil samples

Cooling rate:  $10^{-3} - 10^{-1}^\circ\text{C/sec}$

Supersaturation with respect to water: 0 - 2%

CCN concentration:  $0 - 10^3/\text{cc}$ , and room air

The ice nucleation behaviors of samples will be displayed on the supercooling-supersaturation diagram with other factors as parameters.

#### B.1.8 Instrumentation

Instruments to be used are the modified LaMer-Sinclair Monodispersed Aerosol Generator, a smoke box (Class 13, Instrumentation), an Argon Ion Laser (Off shelf), Spectra-Physics, Model 162), a camera (Off shelf), and the DU-SDI Chamber.

The DU-SDI Chamber, as shown in Figure B-1, consists of two parallel copper plates of the dimension 25 cm x 25 cm x 0.5 cm with one side wall made of copper of the dimension 25 cm x 4 cm x 0.5 cm. The other three walls are made of Plexiglas of the same dimensions. The relative heating of the top plate and cooling of the bottom plate will be performed by thermoelectric modules attached to the edges of plates on the opposite side of the copper wall. The top plate also carries a heat exchanger for circulating liquid coolant. The temperature controls for coolant as well as the power for the thermoelectric modules are made programmable so that the coolant regulates the temperature of the top plate and the power supply controls the temperature difference of  $T_1 - T_2$ . The insides of the top and bottom plates carry wet filter papers to supply moisture. The laser beam will be sent in from the module side of the wall.

#### B.1.8.1 Measurement and Data Requirements

For a given sample air composition, the chamber plate temperature at an arbitrary horizontal position defines the supersaturation distribution on the median plane in the chamber. Photographic data provide the numbers of ice crystals per unit of chamber volume as a function of time and ambient conditions. A recorded commentary will be utilized at appropriate points during the experiment along with digital recording of time, temperatures, and pressure.

#### B.1.8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	3
Type	4
Pollutant	
Pressure	
Temperature	4
Relative humidity	3
Charge	

<u>Parameters</u>	<u>Variations</u>
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### B.1.9 Procedure

General activity details are given below followed by a representative activity time line. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with thought to translating this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure B-2).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<p><u>Procedure 1</u></p> <ul style="list-style-type: none"> <li>• Prepare nuclei in smoke chamber</li> <li>• Purge DU-SDI chamber</li> <li>• Cool chamber to desired temperature <math>T_M (\Delta T = T_1 - T_2 = 0)</math></li> <li>• Establish thermal and vapor equilibria at <math>\Delta T = 0</math></li> <li>• Inject aerosol nuclei, wait until turbulence dies down</li> <li>• Increase <math>\Delta T</math> slowly while holding <math>T_M</math> constant</li> <li>• Record plate temperatures <math>T_1</math> and <math>T_2</math> and time (continuous)</li> <li>• Time-lapse photographs of ice crystal formation</li> <li>• Recycle for new mean temperature (ten values)</li> <li>• Recycle for CCN (two values)</li> <li>• Recycle for various nuclei</li> </ul>	<p>20</p> <p>5</p> <p>10</p> <p>10</p> <p>5</p> <p>30</p> <p></p> <p></p> <p></p> <p></p> <p></p>

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<p><u>Procedure 2</u></p> <ul style="list-style-type: none"> <li>• Prepare nuclei in smoke chamber</li> <li>• Purge DU-SDI chamber</li> <li>• Cool chamber to cloud base temperature <math>T_M (\Delta T = 0)</math></li> <li>• Establish thermal and vapor equilibria at <math>\Delta T = 0</math></li> <li>• Inject aerosol nuclei, wait until turbulence dies down</li> <li>• Increase <math>\Delta T</math> slowly while <math>T_M</math> lowers according to preprogramming</li> <li>• Record plate temperatures <math>T_1</math> and <math>T_2</math>, and time (continuous)</li> <li>• Time-lapse photographs of ice crystal formation</li> <li>• Recycle for new cloud base temperature (four values)</li> <li>• Recycle for CCN</li> <li>• Recycle for various nuclei</li> </ul> <p>The variables to be considered are nuclei types, size distributions, temperature, supersaturation, and fog size and number concentration.</p>	<p>20</p> <p>5</p> <p>10</p> <p>10</p> <p>5</p> <p>30</p>

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<p><u>Procedure 1</u></p> <ul style="list-style-type: none"> <li>• Generate nuclei within smoke chamber Smoke particle number concentration check by ultramicroscope Smoke concentration adjustment by dilution with clean air</li> <li>• Purge DU-SDI chamber</li> <li>• Cool chamber to desired temperature <math>T_M</math> (<math>\Delta T = T_1 - T_2 = 0</math>)</li> <li>• Establish thermal and vapor equilibria at <math>\Delta T = 0</math></li> <li>• Inject aerosol nuclei, wait until turbulence dies down</li> <li>• Increase <math>\Delta T</math> slowly while holding <math>T_M</math> constant</li> <li>• Record plate temperatures <math>T_1</math> and <math>T_2</math>, and time (continuous)</li> <li>• Time-lapse photographs of ice crystal formation</li> <li>• Recycle for new mean temperature <math>T_M</math> (ten values)</li> <li>• Recycle for CCN (three values)</li> <li>• Recycle for various nuclei (four values)</li> </ul>	<p>20</p> <p>5</p> <p>10</p> <p>10</p> <p>5</p> <p>30</p>
<p><u>Procedure 2</u></p> <ul style="list-style-type: none"> <li>• Generate nuclei within smoke chamber</li> <li>• Purge DU-DSI chamber</li> <li>• Cool chamber to cloud base temperature <math>T_M</math> (<math>\Delta T = 0</math>)</li> <li>• Establish thermal and vapor equilibria at <math>\Delta T = 0</math></li> <li>• Inject aerosol nuclei, wait until turbulence dies down</li> <li>• Increase <math>\Delta T</math> slowly while <math>T_M</math> lowers according to preprogramming</li> <li>• Record plate temperatures <math>T_1</math> and <math>T_2</math>, and time (continuous)</li> <li>• Time-lapse photographs of ice crystal formation</li> </ul>	<p>20</p> <p>5</p> <p>10</p> <p>10</p> <p>5</p> <p>30</p>



**PROCEDURE**

<b>DETAILED ACTIVITY</b>	<b>TIME REQUIRED MINUTES</b>
<p><u>Procedure 2 (continued)</u></p> <ul style="list-style-type: none"><li>● Recycle for new cloud base temperature (four values)</li><li>● Recycle for CCN (three values)</li><li>● Recycle for various nuclei (four values)</li></ul>	

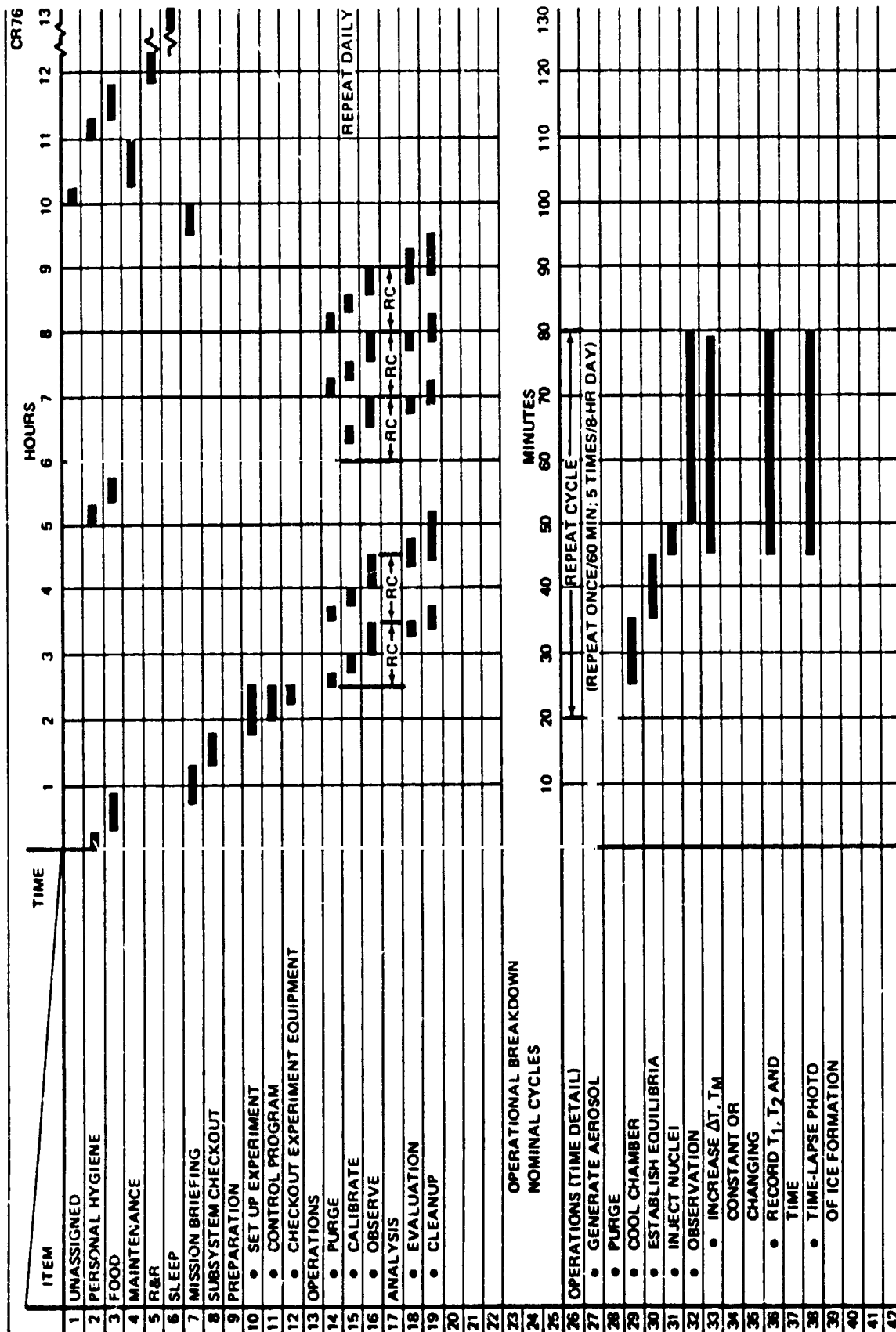


Figure B-2. Activity Timeline (One Day) Experiment Class 2

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## B. 2 ICE NUCLEATION APPROACH 2

### B. 2.1 Introduction

The importance of the ice phase in precipitation processes was first pointed out in Bergeron's classic paper in 1935 and since that time an interest in naturally occurring ice forming nuclei has developed.

There are three basically different methods by which ice forming nuclei become effective in forming ice particles in clouds. The relative importance of these three mechanisms namely, sublimation, bulk freezing and contact can be very well studied in a zero-g laboratory.

Supercooled drop freezing by contact nucleation has been observed and reported during the course of the studies of ice nucleation in our laboratories (N. Gokhale and J. Goold, Jr., "Droplet freezing by Surface Nucleation", J. APPL. MET., vol. 7 1968; N. Gokhale and O. Lewinter, "Microcinematographic studies of contact nucleation", J. APPL. MET., vol., 10, 1971; N. Gokhale and J. Spengler, "Freezing of freely suspended, supercooled drops by contact nucleation", J. APPL. MET., vol., 11, 1972). The drops were either supported on a metal plate or were freely suspended in a vertical wind tunnel. Silver iodide particles were effective in freezing millimeter size drops at -4 to -5C. Particles of naturally occurring silicates were found to be effective in the range -7 to -10C. Thus the ice nucleating ability of such dry particles is much greater by the contact mechanism than when particles are embedded in the drops. The importance of this finding in forming the solid hydrometeors in cumulus clouds is discussed in the above mentioned papers.

It is suggested to carry out similar experiments in cold chambers under zero-g conditions so that the drop and the particle distribution could be varied over a wider range.

#### **B. 2. 2 Objective**

Determine the absolute nucleating efficiencies as a function of nuclei types, size distributions and relative humidities. In addition, study the relative importance of sublimation, bulk freezing and contact nucleation of ice.

#### **B. 2. 3 Scientific Justification**

At present it is not definitely known how ice forming nuclei become effective in a growing cumulus cloud. It is essential to know the nucleation mechanism and conditions for optimum nucleation effectiveness and how it depends on the size and type of nuclei.

#### **B. 2. 4 Applications**

These data are important to effectively seed a cold cloud to increase or to redistribute the precipitation such as snow, hail and cold fogs. These studies would pinpoint the effective type of seeding material in the precipitation cycle and how much material is required to achieve the desired result.

#### **B. 2. 5 Terrestrial Laboratory Limitations**

The earth bound cloud physics laboratories face a number of restrictions in their experiments due to the suspension systems, supports, convection, temperature and humidity variations in the chamber and fall out due to gravitational forces. The comparative studies to determine the effectiveness of different nucleation modes, such as sublimation, condensation and freezing and contact, are difficult to carry out under limitations of a terrestrial laboratory.

#### **B. 2. 6 Zero-Gravity Opportunities**

When ice nucleation takes place in the presence of a gravitational field, ice particles formed move away from nucleation sites and hence it is difficult to make a conclusive analysis of nucleation modes. The zero-gravity environment in a space laboratory would be most appropriate to perform experiments to answer the questions as to the effective surface properties and size distributions of nuclei to form ice crystals at comparatively warmer temperatures.

### B.2.7 Quantification

It is extremely difficult or nearly impossible to accurately predict the progress and success of many scientific experiments. However, as discussed in the earlier section, the proposed experiments would be best carried out in a zero-g field.

### B.2.8 Approach

#### B.2.8.1 General

When the concentration of natural ice-forming nuclei is not sufficient in supercooled clouds, introduction of artificial ice nuclei helps to initiate the thermodynamic change and modifies the cloud structure, often leading to additional precipitation.

In order to modify clouds, we must know the complex processes of ice nucleation, both on the micro and macro scales. The microphysical processes of nucleation such as sublimation, condensation-freezing and contact depend on the environmental water vapor saturation ratios with respect to ice and liquid surfaces.

The freezing of supercooled drops by contact nucleation at very warm temperatures had been first reported by us in 1968. Since then this effect has been well confirmed by other laboratories. The mode of nucleation by contact is most effective in forming ice particles at comparatively warmer temperatures than those possible by other ice nucleating mechanisms. Under zero-g conditions, suspension systems, supports and fall out can be avoided and hence the contact mechanism can be better studied for its effectiveness.

The method will consist of introducing ice-nuclei in the Static Thermal Diffusion Ice chamber (SDI) maintained at subfreezing temperatures. The chamber plate temperatures and pressure define the relative humidity distribution in the chamber. A laser beam and photographic techniques will be used to count the ice crystals formed per unit volume as a function of time and ambient conditions. The saturation ratio in the chamber can be

maintained over a wide range, from ice saturation to super saturation with respect to water. The temperature in the chamber will be varied over a wide range and crystal counts will be determined. Absolute nucleation efficiencies will be studied as a function of nuclei types (inorganic, organic, soil samples and artificial nuclei like AgI, AgI-NaI, PbI<sub>2</sub>, CuS) and size.

#### B.2.8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	
Type	6
Pollutant	4
Pressure	3
Temperature	4
Relative humidity	4
Charge	3
Rate of cooling	
Time	6
Sound	
Electric field	3
Nuclear radiation	3
Adsorption	4
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	3
Surface tension	
Aerosol age	3



<u>Parameters</u>	<u>Variations</u>
History	3
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### B.2.9 Procedure

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure B-3).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<u>Preparation</u>	
• Set up experiment	30
• Control program	15
• Check out equipment	15
<u>Operations</u>	
• Preparation of nuclei in conditioning chamber	10
• Purge diffusion chamber	5
• Establish thermal equilibrium	20
• Inject nuclei	2
• Cool chamber to desired temperature	20-40
• Continuous record of temperature and time	
• Time-lapse photographs of ice-crystal formation	
• Recycle for new temperature (cooling rate 0.25°C/min).	
• Recycle for various nuclei	
<u>Analysis and Checkup</u>	
• Data evaluation	40
• Cleanup and Shutdown	20

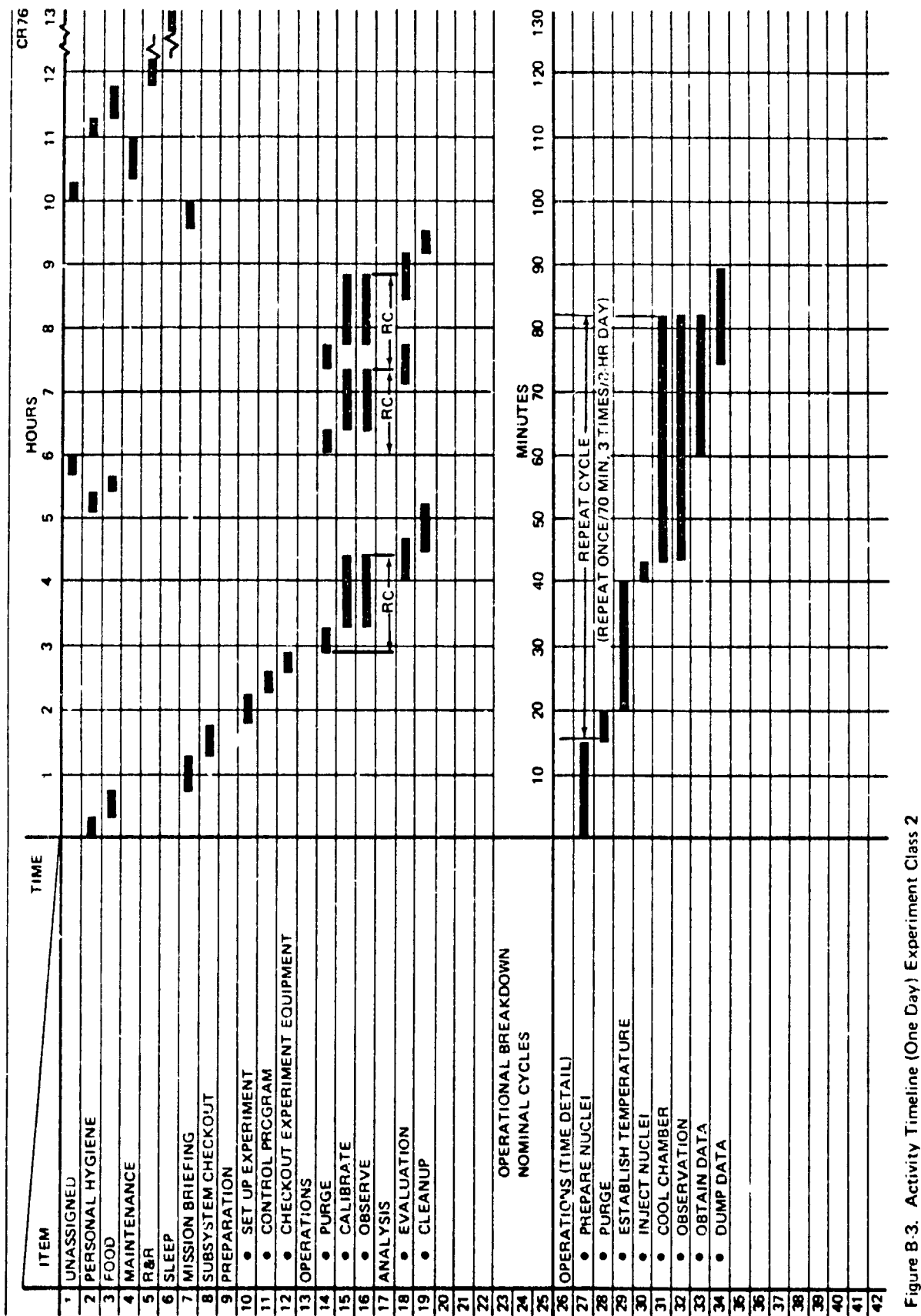


Figure B-3. Activity Timeline (One Day) Experiment Class 2

### B.3 ICE NUCLEATION APPROACH 3

#### B.3.1 Introduction

Serious concern is being expressed concerning the inadequacy of our knowledge about ice formation processes in the atmosphere. A detailed knowledge of the mechanisms of ice formation lies at the heart of understanding thunderstorm precipitation, hail formation and suppression, and charge separation in glaciated cloud. Our most prominent means of weather modification depends upon the concept that nature is unable to supply adequate numbers of ice nuclei which are active at relatively low degrees of supercooling, and that the injection of artificial ice nuclei will have the desired effect in either promoting precipitation or in suppressing hail.

Comparison of the different techniques which have been devised for measuring ice nuclei concentrations have shown such wide disparity that field programs have largely adopted an operational point of view, deciding to ignore measurements of the concentration of natural ice nuclei and proceed using statistical analyses of the results of randomized experiments in order to evaluate the results of field seeding operations.

The reasons for the slow rate of development of an understanding of the physics of ice nucleation in the atmosphere is not altogether clear but part of the reason must be closely related to inadequacies in laboratory facilities for simulating the manner in which a supercooled cloud begins to glaciate. Moreover, a much too simplistic point of view has been taken concerning the interaction of ice nuclei and cloud dynamics.

There appears to be emerging at least the rudiments of a consensus that ice in a cloud is produced by two main processes, namely (1) deposition and (2) freezing. The freezing process is further complicated by the different mechanisms through which the particle containing some ice nucleating capacity enters the droplet: (1) direct condensation in warm clouds as the result of the nucleus simultaneously possessing characteristics

common to both ice nuclei and condensation nuclei and (2) scavenging of the nucleus by either a warm or supercooled cloud. In both the condensation process and warm cloud scavenging mechanism, the ice nucleus is likely to be deactivated or have its ice nucleating properties greatly altered. The larger ice nuclei are expected to be most likely to serve as condensation nuclei while the smaller ice nuclei are more likely to be scavenged since their Brownian diffusion coefficient is higher.

The number of ice nucleating events depends upon a host of cloud physical parameters which are not easily duplicated by simple ice nucleus counting techniques. It now seems unlikely that a single ice nucleus counter will suffice to evaluate the ice nucleating capability of the natural aerosol under all cloud conditions.

A better understanding of the microphysics of ice nucleation is badly needed in order to enable the scientific community to develop a procedure for evaluating the ice nucleating potential of the natural aerosol. In a zero-g environment, long cloud confinement times, comparable to the lifetime of droplets in natural cloud, can be obtained so that scavenging and deactivation mechanisms can operate on a time scale similar to that which occurs in a natural cloud.

### B.3.2 Objective

The objective of this experiment is to simulate both the range of thermodynamic conditions and the time rate of events in natural cloud in a cooled wall expansion chamber in an effort to determine what microphysical processes are important in atmospheric ice nucleation. These studies are essential to evolving a simple field technique for measuring or evaluating the number of nuclei possessing ice nucleating characteristics which are effective in the production of ice in cloud under a given set of circumstances.

### B. 3. 3 Scientific Justification

The whole concept of weather modification implies that man can intervene in the natural course of weather processes by altering some parameter which exerts a sensible influence on the rate of release of large quantities of energy. Fortunately, it is just those phase transitions of water and the accompanying energy releases that offer a throttle [to use the words of Braham (1)] which can be manipulated through the introduction of artificial nucleating agents in order to accomplish a measure of intervention (2). It is the complex interaction between the rate of energy release and the role it plays in driving the hydrodynamic motions that is beginning to be understood through numerical modeling. Almost every cloud microphysical phenomenon plays some role in regulating the liquid water content, in determining the distribution of water between the liquid and solid phase, and/or in influencing the nature of the precipitation. Some of these processes are critically dependent upon a variety of forms of heterogeneous nucleation. So far, the microphysical processes have been incorporated in the numerical models in only very rudimentary forms (3). This is perhaps all that is warranted, considering the state of confusion which surrounds such important processes as ice nucleation (4).

Those atmospheric nuclei which are taken into the cloud base initially may experience the environment characteristic of warm cloud development for which a slight degree of supersaturation exists in the region of cloud base. A certain percentage of the ice nuclei are capable of acting either as condensation nuclei or as ice nuclei depending on the exact environment (warm or cold cloud) to which they are subjected first. Nuclei with "potential" ice nucleating activity which first pass through the base of warm cloud may be deactivated or their ice nucleating activity greatly altered by serving as condensation nuclei (18). Several factors may govern the actual percentage which will be lost through this process. First, the nuclei distribution itself is a factor since, in general, larger nuclei are more efficient as condensation nuclei; second, the maximum supersaturation achieved and the length of time the nuclei are exposed to it are important since both of these factors increase the probability that smaller nuclei will be activated as centers of condensation.

Those nuclei which are not deactivated by serving as condensation nuclei may still be lost before reaching the freezing level by warm cloud scavenging. The percentage of ice nuclei lost through in-cloud scavenging depends on the time spent in the warm cloud, the mass and size distribution of the "potential" ice nuclei, and the concentration and size distribution of the cloud droplets.

If the ice nuclei have passed the freezing level (having escaped serving as condensation nuclei or being scavenged) and become supercooled, they may serve as either sublimation nuclei or contact freezing nuclei. The contact nucleation process is again dependent on the scavenging mechanism so that freezing depends upon the product of the scavenging efficiency and the probability that the coagulation event will induce freezing. The latter is probably highly dependent upon the degree of supercooling, characteristic of ice nucleation thresholds and the degree of supersaturation with respect to ice. In addition, the insoluble nuclei which were scavenged by water droplets under warm cloud conditions or served as condensation nuclei may cause the droplet to freeze as it supercools. However, the freezing activity of these ice nuclei may bear no resemblance to the deposition or sublimation activity had the nucleus remained essentially dry until injected into a supercooled environment (5) and may depend very strongly upon whether the nucleus is completely immersed in the droplet or left projecting from the surface as a result of a hydrophobic region on its surface (19). Thus, there are three separate processes by which the ice nuclei can initiate the formation of ice particles: sublimation, contact freezing, and internal or surface freezing. There is substantial evidence that all of the processes do in fact occur in nature. The various ice nuclei counting techniques tend to emphasize one or more of these processes while de-emphasizing the others, thus greatly distorting the ice nucleating potential to be expected in real cloud.

The weight of evidence indicates that ice nuclei may act in a number of modes which differ considerably in their cloud physical aspects. There is a real need to study these different modes of ice nucleation in detail both separately and as they may act in concert in clouds in order to verify the correctness

of these various hypotheses. The nucleation "thresholds"\* for each of these various modes of nucleation need to be studied with an apparatus which can reproduce cloud conditions quite accurately to meet the needs of the experiment. The UMR cooled wall type cloud simulation chamber (adapted to function in Zero-g) is specifically designed to meet these requirements. However, such a complex facility is not useful as an operational ice nucleus counter for field observations, so another approach must be sought to satisfy the latter need.

It would seem that, due to the complex behavior of ice nuclei and the variety of modes in which they may nucleate ice, several different types of ice nucleus counters should be employed simultaneously. For instance, one method of ice nucleus counting should be adapted which almost exclusively measures the efficiency of contact nucleation where the type of cloud is well known. Another should be employed which measures the efficiency of sublimation or deposition nucleation. One counter should measure the condensation nucleus characteristics of the aerosol. From this information and a knowledge of the cloud physics, models might be developed which can predict the total number of ice nucleating events resulting from all the different modes for a given set of cloud conditions. There will be cases where experience will indicate that only one type of measurement need be made; perhaps most of the nucleation will result from contact nucleation.

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\*We hesitate to employ the word "threshold" because it implies that one reaches a given degree of supercooling at which point all nuclei with the same "activity" suddenly nucleate ice. Heterogeneous nucleation is a probabilistic process where for identical nuclei exposed to a given set of thermodynamic conditions there is a certain probability per unit time per nucleus that nucleation will be induced in the case of deposition or freezing nucleation. However, in the case of heterogeneous nucleation, it has not been possible to produce nuclei with identical characteristics and one is always constrained to study a collection of nuclei with a distribution of nucleation probabilities. Under these circumstances the term "threshold" still conveys an operational definition which is useful.



For instance, if cold clouds are being seeded from aircraft, perhaps contact nucleation will play a dominating role and only this type of ice nucleus counter will be needed to evaluate the seeding agent. However, if the circumstances of the cloud physics are such that cloud base is quite warm, in order to evaluate whether a deficiency of natural freezing nuclei exists, perhaps all three types of ice nucleus counters should be employed. Circumstances and experience must serve as a guide. Until the physics, chemistry, and meteorology of ice nuclei are better understood, it would be better to be on the safe side and make too many measurements. Later the number of measurements can be tailored to suit the situation.

In the U.S. National Academy of Sciences report, Weather and Climate Modification - Problems and Progress (1973), the statement is made:

"Although much work has been done to develop a reliable technique for the detection and counting of ice nuclei in the air, a satisfactory method that counts ice nuclei under conditions that approximate closely those in natural clouds is not yet available."

#### B. 3.4 Applications

At this time the scientific community has no technique which can be relied upon to yield the concentration of ice nuclei which are effective at any given degree of supercooling and supersaturation with respect to ice. The knowledge developed from the kinds of simulation experiments proposed here will be valuable in developing a new field technique for estimating the numbers of ice nuclei in the atmosphere which are capable of responding as ice nuclei under the conditions which exist in natural cloud. This problem is given a very high priority by both the National Academy of Sciences panel on Weather and Climate Modification and the Committee on Nucleation of the International Commission on Cloud Physics.

"To repeat, the emphasis that has been given to the manner in which AgI nucleates ice is not based on assurance that the present findings are conclusive but rather to illustrate how uncertain we are in our understanding of this basic problem and great is the practical value of definitive knowledge on this subject. We must recognize that we are still attempting cloud modification in a state of incomplete knowledge of essential processes; and it is reasonable to believe that future

improvements in such basic knowledge will markedly lower the degree of empiricism of our efforts. "

A more complete understanding of the ice nucleation process and determination of the most effective mode of nucleation will give researchers the information needed to tailor the aerosol for optimum effectiveness.

Obviously, a better understanding of ice nucleation will promote much better cloud modeling procedures in addition to improving our interpretation of cloud physics observational data which is being taken with current instrumentation. With improvement in cloud observations of the turbulent fields using doppler radar and inertial platform techniques, a more detailed knowledge of the factors which play prominent roles in determining the efficiency of ice nucleation in cloud should assist the scientific community greatly in unravelling the interplay or interaction between cloud dynamics and cloud microphysics. The latter is sure to receive high priority as a research objective throughout the next decade.

#### B. 3. 5 Terrestrial Laboratory Limitations

Ice nucleation most often takes place in the presence of supercooled cloud droplets which establishes the supersaturation with respect to ice. It is generally believed that contact between the ice nuclei and supercooled water droplets is one of the most efficient ice nucleating mechanisms, whereupon the scavenging of the nuclei by cloud droplets as a result of Brownian diffusion of the aerosol particles becomes an important factor. Since diffusion is the time dependent process, time is required for this mechanism to be effective. The retention time of laboratory apparatus for cloud is limited by the terminal velocity of the droplets under the influence of gravity and the height of the chamber. Although chambers with large height can be built for terrestrial laboratory use, it becomes increasingly difficult to maintain careful control over the thermodynamic conditions throughout large chambers and it is difficult to provide uniformity of aerosol content throughout large volumes. Since ice particles grow rapidly under the conditions of ice supersaturation provided by water saturation at the same temperature, the ice crystals fall out of the volume rapidly.

Since the nucleated crystals rarely have time to grow to an easily distinguishable size, the crystals have customarily been allowed to fall into sugar solution where they continue to grow to some easily recognizable size. Where longer periods of time are desired, the sugar replicating device is no longer feasible because the crystals falling into the liquid first have ample time to become excessively large. The times required for simulating contact nucleation are not available in the terrestrial laboratory facilities.

#### B. 3.6 Zero-Gravity Opportunities

Since ice nucleation obviously takes place by several different mechanisms, simulating the time rates of events as well as the thermodynamic conditions is essential. While the thermodynamic conditions and cloud environment can be readily simulated in the terrestrial laboratory, the time variable cannot be suitably extended to times commensurate with what is found in nature so that the effect of this variable has been difficult if not impossible to explore. In zero-g, particles which fall out under the influence of gravity are eliminated. The relative positions of nuclei, droplets and ice crystals remain fixed. In effect we have a closed parcel available for study. No longer will the experimenter have to rely upon some method of replicating the ice crystals which fall out of the volume; they will have ample time to grow to an easily distinguishable size in situ. Moreover, the rates of growth can be studied for both the evaporation of the supercooled cloud and for the developing ice crystal population if suitable means of detecting and distinguishing the two populations can be developed.

#### B. 3.7 Quantification

Currently no technique has been available where ice nuclei can be subjected to conditions similar to those found in natural clouds, including the time scale of events. As a result, the nuclei population observed in ice nucleus counters does not represent the same portion of the spectrum which is effective in producing ice in clouds. The most difficult part to simulate quantitatively has been the contribution due to contact nucleation where the time scale is extremely important. Since a given nucleus with ice nucleating characteristics may be able to nucleate ice by several different mechanisms in addition to possessing

some condensation nucleus characteristics, the life history of the nucleus is extremely important. By being able to vary the time spent in the warm cloud regime and the cold regime, these dependent characteristics of the nucleus spectrum can be determined.

It seems to be easily possible to photograph the cloud through a polaroid where the cloud is illuminated with polarized light in order to independently resolve the ice portion of the cloud. It seems highly probable that these experiments can go a long way toward resolving the questions relative to ice nucleation processes.

#### B. 3. 8 Computer Data Acquisition and Control System

The precision, speed, and complexity of the simulation chamber project necessitate an automated data acquisition and control system. The simulation chamber and its supporting peripheral equipment contain over 100 data sensors which measure temperatures, flow rates, motor speeds, laser scattering signals, etc. These measurements have to be collected, converted to digital form, stored, displayed in a form useful to the operator, and used as feedback to control the experiment. Control signals are needed for opening and closing valves, turning pumps on and off, adjusting the speed of the expansion system piston motor, etc.

One experiment with all of its preparation will take several days or perhaps weeks. While the zero-g chamber will be capable of performing an experiment in a much shorter time, the rate of data acquisition increases so that the need for computer control is still required. During the preparatory phase the various support peripherals such as the air preparation system and the aerosol generator are brought into operation and allowed to come to equilibrium. These systems and their products need to be monitored periodically so that their equilibrium can be identified when it occurs and the next phase of the project begun. The computer system can collect all of the relevant data, combine various pieces of data and process it when necessary, and then display a relevant result to the operator who can judge when he is satisfied that equilibrium has been obtained. Then he can tell the computer to continue with the next phase of the project.

When all of the preparations are completed and the actual expansion is begun, the number of data channels which need to be monitored goes down substantially, but those which remain must be monitored at a much faster rate. The chamber wall temperature, the gas pressure, and the laser scattering signals are the crucial data needed to control and analyze the experiment. The most important task is keeping the chamber walls and the gas at the same temperature during the expansion. The thermo-electric modules are pre-programmed to cool the walls according to a given schedule. The task of the control system is to cool the gas in such a way that the gas temperature always equals the wall temperature. The gas temperature is controlled rather than the wall temperature because the gas has a much faster response time to the control signal.

The wall temperature is measured directly with a transistor thermometer. The gas temperature is measured indirectly. The gas pressure is measured with a transducer, and the gas temperature is calculated from this via the ideal gas law. This result is then corrected for the latent heat from condensing droplets. The data from the laser measurements is used to determine this liquid water content. The minicomputer is fast enough to do all of the calculations in a time that is short when compared to the control response time of the simulation chamber.

Approach 3 - Attachment A  
ESTIMATE OF DISTURBING INFLUENCE OF DIAGNOSTIC  
LIGHT BEAMS ON CLOUD PHYSICS EXPERIMENTS

Light beams (whether incandescent, flash, or laser), which are used to observe the phenomena under investigation, can exert a disturbing influence on the system. Laser illumination is preferred because it is monochromatic, polarized, and nearly perfectly collimated. The chief influence will be due to the heating of the bulk gas and/or water droplets under investigation. The following analysis estimates these heating effects.

The Effect of Illumination on the Droplet and Gas Temperature

The rise in the internal temperature of a water droplet illuminated by a low intensity beam with wavelength in the visible is negligible. For a range of realistic conditions, the rise in temperature is of the order of  $10^{-5}$  °K/sec. To illustrate this, we choose the following set of assumptions:

He-Ne laser wavelength:	$\lambda = .6328 \mu$
power output of laser:	$P_0 = 10^{-1}$ joules/sec (100 m watts)
radius of laser beam:	$R = 0.85$ cm
imaginary part of index of refraction of water at $\lambda = .6328 \mu$ : *	$\text{Im}(n) = 1.6 \times 10^{-8}$
absorption coefficient for water:	$\alpha = \frac{4\pi \text{Im}(n)}{\lambda} = 3.2 \times 10^{-3}$ /cm
radius of the water droplet:	$r = 10^{-3}$ cm (10 $\mu$ )
specific heat of water:	$c = 1$ cal/gm/°K
density of water:	$\rho = 1$ gm/cm <sup>3</sup>

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\*The imaginary part of the index of refraction for water at .6328  $\mu$  and 25°C was taken from George M. Hale and Marvin R. Querry, 'Applied Optics, 12, 555 (1973).

The rise in the internal temperature of the water droplet† (when  $ar \ll 1$ ) is given by

$$\begin{aligned}\Delta T &\approx \frac{P_0}{4.186} \frac{r^2}{R^2} \left(\frac{ar\pi}{4}\right) \left(\frac{4}{3}\pi r^3 \rho\right)^{-1} \cdot \text{K/sec} \\ &= 4.5 \times 10^{-2} \frac{P_0 a}{R^2} \\ &= 2 \times 10^{-5} \cdot \text{K/sec}\end{aligned}\tag{1}$$

As can be seen from Eq (1),  $\Delta T$  is independent of the droplet radius. This result is dependent on the condition  $ar \ll 1$ .

The rise in temperature of the gas illuminated by the 100 m watt He-Ne laser is also negligible. For a typical illuminated volume of gas composed of air, saturated water vapor, and aerosols, we can estimate an upper bound on  $\Delta T$  of about  $10^{-5} \cdot \text{K/sec}$ . To do this, we use a total attenuation coefficient (including both elastic and inelastic scattering) which was obtained from experimental data on air with water vapor and naturally occurring aerosols. \*\*

total absorption coefficient:	$a_g \approx 7 \times 10^{-7} \text{ cm}^{-1}$
specific heat of the gas:	$c_p \approx 0.24 \text{ cal/gm} \cdot ^\circ\text{K}$
density of the gas:	$\rho_g \approx 1.3 \times 10^{-3} \text{ gm/cm}^3$
volume of gas:	$\pi R^2 l$

The rise in temperature (when  $al \ll 1$ ) is given by

$$\begin{aligned}\Delta T &\approx \frac{P_0 a_g l}{4.186 c_p \pi R^2 l \rho_g} \\ &= 2.4 \times 10^{-2} \frac{P_0 a_g}{R^2} \\ \Delta T &\approx 2 \times 10^{-5} \cdot \text{K/sec}\end{aligned}\tag{2}$$

†The quantities in Eq (1) have the units given in the definitions. The units in parentheses are explanatory.

\*\*This value of  $a_g$  was taken from L. Elterman, Air Force Cambridge Research Laboratory, report 0155 (1968), p. 34, for an altitude of about 1 km and 15 C.

The total attenuation coefficient was taken from an average of seven different experiments at an altitude of 1 km for real atmospheric conditions. A rough estimate for the corresponding concentration of  $2\ \mu$  aerosol particles is about  $100 \pm 50$  particles per  $\text{cm}^3$ . The  $\alpha$  we have used includes the elastic Rayleigh scattering attenuation coefficient and also the effect of any water vapor (saturated or otherwise) which is present. Under most conditions, the experiments were not performed on water saturated air. However, the effect of the water vapor will be small. At 100 percent relative humidity, water clusters will have nucleated only on the impurity aerosols and are in effect no more dense than the aerosol particles themselves.

Subject to the above considerations, we can limit the rise in temperature of the illuminated gas to no more than  $10^{-5}\ ^\circ\text{K}/\text{sec}$ . Thus the heating effect of the  $.6328\ \mu$  laser beam will be negligible for both the gas and the water droplets.



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Appendix C  
CLASS 3  
ICE MULTIPLICATION

It is well established that the number of ice crystals in certain clouds such as cumulus clouds far exceed that which can be expected from the ice nuclei concentrations at the cloud top temperatures (see for example, Mossop et al., 1970, 1972, Auer et al., 1969, Hobbs 1969). There is no satisfactory explanation so far to account for this discrepancy but it seems likely that large drops or large graupel particles are necessary before this ice multiplication process occurs. A considerable number of experiments have been made to test the hypothesis that ice multiplication takes place either through the shattering of large freezing drops or during the process of graupel formation by riming or both. Unfortunately so far no laboratory experiment has yielded satisfactory evidence to suggest that either of these mechanisms is correct.

One of the probable causes for the failure of laboratory experiments may be the inability of observing the freezing of drops over a large period of time except by using a suspension device.

Early experiments on drop freezing were performed in Mason's Laboratory at University of London. A description of these experiments which were performed with drops suspended between immiscible liquids or suspended in fine fibers are described by Mason (1972). Early results showing fragmentation of drops were attributed by Dye and Hobbs (1966) to have been caused by the presence of  $\text{CO}_2$  in their apparatus. Apart from the presence of  $\text{CO}_2$  the experiments of Johnson and Hallett (1968) and Dye and Hobbs (1968) further showed that shattering of drops was largely due to their being not in thermal and solution equilibrium with the environment. In trying to simulate in cloud conditions as best as one could, Kuhns (1966), Hobbs and Alkezweeney (1968), Brownscombe and Thorndike (1968) performed freezing of drops in

free fall. Unfortunately the results of these experiments were not consistent enough to establish any reliable mechanism or statistics on the production of splinters by freezing of drops.

It is expected that the understanding of the multiplication mechanism may be achieved through understanding of the freezing process of a drop. Careful observation of a freely suspended drop during freezing will produce useful information on the freezing process.

### C.1 OBJECTIVE

The specific purpose of this experiment is to determine the extent to which ice fragments are formed during the interaction of a super cooled water drop with an ice crystal i. e., during the freezing of water drops. This objective may be achieved through the detailed observation of the freezing process of the water drop freely suspended in thermal and solution equilibrium with the environment.

### C.2 SCIENTIFIC JUSTIFICATION

The philosophy of weather modification is based on the fact that there are insufficient ice crystals in a cloud for the release of precipitation. Mason (1955) showed that the number of ice crystals must be  $10^2$  to  $10^3$  times higher than that of the ice nuclei concentration at -15 C. This deficiency may therefore be rectified by adding artificial ice nuclei. On the other hand ice crystal concentration measured in certain cumulus clouds indicate that certain natural processes are existing in these clouds to generate ice crystals far in excess than those expected. This raises the fundamental question as to the necessity of knowing which clouds and the condition that are conducive to the production of secondary ice crystals so as to make a decision as to seed or not. Without this knowledge it will not be possible to know whether a cloud requires seeding to make it rain and how much seeding material is required. Furthermore the danger of overseeding a cloud thereby reducing the chances of precipitation, may be avoided if rain augmentation is the objective or encouraged if reduction of rainfall is required.

### C.3 APPLICATIONS

- A. Weather Modification
- B. Numerical cloud modelling
- C. Severe weather forecasts

### C.4 TERRESTRIAL LABORATORY LIMITATIONS

Terrestrial experiments on ice multiplication are limited by the time of observation of the freezing drop. Therefore in order to study the detailed freezing process it is required to use substrates or suspension systems. Both these systems interfere with the heat transfer during freezing thereby modifying the freezing process. Wind tunnels are useful in this aspect but the control of a freezing drop within the wind tunnel during the freezing is difficult due to shift of the center of mass. Effects such as rotation, oscillation and shifting are observed with the tunnel experiments.

### C.5 ZERO-GRAVITY OPPORTUNITIES

An important aspect of ice multiplication is to understand in detail the freezing process of a drop. In order to do this the drop must be freely suspended in air without any support and observed for the whole period of freezing. The zero-gravity laboratory will eliminate the laboratory restriction of observation time or use of wind tunnels. It must be mentioned that under natural conditions (in cloud), the freezing process is influenced by the ventilation as it falls in a gravity field. The influence of ventilation is to make the heat transfer across the drop non-symmetric. With the zero-gravity one is restricted only to symmetric heat transfer. But this information is essential to understand the mechanism of freezing and the cause for any shattering that may occur. At a later stage correction for ventilation may be achieved.

### C.6 QUANTIFICATION

As evident from the previous discussions, after almost two decades of laboratory experiments our understanding of the freezing process of a drop is not clear. By observing a single drop for a longer time under controlled conditions in a zero-gravity environment we should be in a position to obtain considerable insight into mechanism such as distribution of bubbles in the freezing drop, the direction and rate of freezing and any ejection of ice crystals.

## C.7 APPROACH

### C.7.1 Chamber

This experiment requires a static diffusion chamber or an expansion chamber. With the SDI, control of temperature and humidity may be done via an external source. Apart from this the operation procedure is the same for both.

Although the advantages of both these chambers are very much the same, the SDI is preferred for this experiment because of the smaller size of SDI. Achievement of the environmental properties may be easier and better controlled with this chamber. Furthermore only one drop need be considered at a time, hence can be maintained at a given location where humidity and temperature is precisely known.

### C.7.2 Controls and Equipment

The controls necessary are

- (1) Temperature
- (2) Pressure
- (3) Dew point or relative humidity
- (4) Gas
- (5) Optical Environment
- (6) Ice particle generator
- (7) Data management and interface electronics
- (8) Miscellaneous support
- (9) Power control
- (10) Console
- (11) Optical and imagery devices
- (12) Tape recorder

### C.7.3 Data Gathering

- (1) Time lapse camera
- (2) IR microscope for temperature sensor
- (3) Vocal record of observation.

This experiment is primarily concerned with observations on the freezing behavior of a drop rather than the measurement of any result, except for the

counting of any secondary ice crystals. Therefore the important aspects are control and measurement of the input parameters such as environmental conditions and drop size and temperature. Photographic record is more than sufficient to obtain useful information for further analysis.

#### C. 7. 4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	4
Type	2
Pollutant	
Pressure	4
Temperature	4
Relative humidity	6
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	

<u>Parameters</u>	<u>Variations</u>
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### C.8 PROCEDURE

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure C-1).



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Unassigned	12
● Personal hygiene	18
● Food	30
● Maintenance	48
● R & R	48
● Sleep	480
● Mission briefing	30
● Subsystem checkout	30
● Preparation	
● Set up experiment	42
● Control program	18
● Check out experiment equipment	18
● Operations	
● Purge differential chamber	1/2 p/ev
● Calibration	5
● Observation	3 per/ev
● Data evaluation	45
● Clean up and shut down	10
● Operations breakdown (Time detail)	
● Purge	5
● Establish temp	10
● Establish RH	10
● Establish pressure	2

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Insert drop</li> <li>• Nucleate drop</li> <li>• Observe freezing and photograph</li> <li>• Allow crystals to grow</li> <li>• Obtain data</li> <li>• Data dump</li> </ul>	<div>1</div> <div>2</div> <div>5</div> <div>1</div> <div>0.5</div>

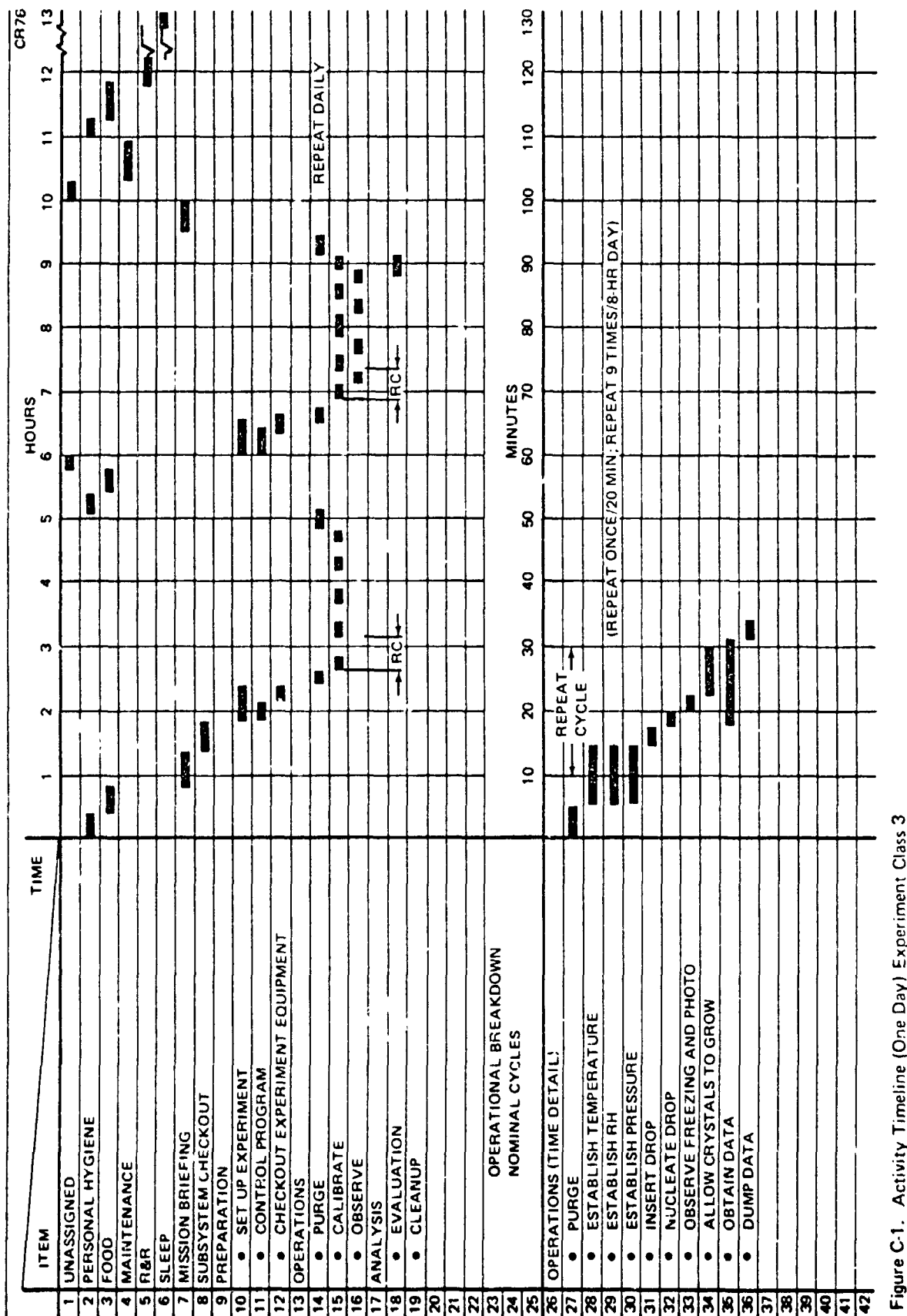


Figure C-1. Activity Timeline (One Day) Experiment Class 3

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Appendix D  
CLASS 4  
CHARGE SEPARATION  
(ELECTRIFICATION)

D.1 INTRODUCTION

Lightning causes much damage to forests, buildings, and homes through fires. Also of concern is the direct loss of life from lightning strikes and the ever present threat of such loss from strikes on aircraft and space vehicle launches. An increased emphasis has been placed on finding what mechanisms are important to the electrification processes within thunderstorm clouds.

A number of interactions involving ice and droplets have been shown to produce varying degrees of particle electrification. Among these interactions are freezing of supercooled water drops, melting of snowflakes and hailstones, the disintegration of large raindrops, and collisions between ice crystals, water droplets and hail pellets. An understanding of the relative importance of these processes with respect to particle electrification is important if any attempt is to be made to minimize lightning conditions.

These electrification phenomena involve a number of very different mechanisms of charge transfer (e. g., by ion segregation at the ice-water interface during freezing, by differential migration of positive and negative ions along a temperature gradient in ice by conduction of charge between ice crystals and water drops colliding with and rebounding from hail pellets, and by the shearing of the electrical double layer at the surfaces of air bubbles bursting in water and of water drops bursting in air).

Most of the above processes occur for hydrometeors (ice, water) which are greater than  $100\text{ }\mu\text{m}$  in diameter. Thus, the physical separation of the charge once electrification has taken place is a function of gravity and convective updrafts within a cloud. But a number of electrification processes

themselves, which are of concern here, are a function of thermal and electrical properties of the particles, which in turn are not a function of gravity. As a consequence of the ice and droplet particle size and resulting fallout due to gravity, most terrestrial laboratory experiments have utilized mechanical supports to provide sufficient time for the necessary experimental observations. The conductivity of even the best supports modifies the resulting charge measurements to such an extent that qualitative measurements are obtained, but quantitative measurements are nearly impossible.

#### D.2 OBJECTIVE

Determine quantitative values for charge transfer occurring during several important atmospheric processes.

#### D.3 APPLICATIONS

Particle-to-particle charge transfer mechanisms are believed to be responsible for the production of charge separation in thunderstorm clouds that results in lightning. An understanding of these mechanisms may permit certain weather control efforts to minimize lightning and associated fires and strike damage to buildings, forests, and aircraft. This understanding will aid in deciding "where" in the cloud system and "when" in the cloud life to modify weather during lightning-associated precipitation.

#### D.4 ZERO-GRAVITY OPPORTUNITIES

Under low-gravity conditions, the various interactions can be studied without the need for physical supports. Techniques are presently available for the measurement of charge on a freely floating spherical droplet by the use of static and alternating electric fields. A low-gravity environment would permit quantitative measurements to be made for those electrification processes which are not strictly gravity-dependent. Even certain aspects of the processes such as electrification during the disintegration of large droplets could be studied to advantage under such conditions.

## D.5 APPROACH

### D.5.1 General

Electrification during the impingement and rebounding of cloud droplet with hail pellets will be used as a representative example of this class of experiments.

A neutrally charged, 1-cm ice pellet will be placed near the center of a thermally controlled, static ice diffusion chamber. The plate temperatures of the chamber will define the relative humidity within the chamber from exact saturation with respect to ice up to several percent supersaturation as required. Humidity values below 100 percent can be provided by utilizing dry walls and plates and preconditioning the air before entry into the chamber.

A uniform electric field will be applied across the diffusion chamber. This field will electrically polarize the ice pellet as would happen in a natural cloud.

Single droplets between 100 to 1,000  $\mu\text{m}$  in diameter will then be impinged at various angles onto the ice pellet so that the droplets will rebound from the surface. The motion of the droplet (and of the ice pellet to a lesser degree due to its mass) under the influence of a static electric field or an applied alternating field will be used to deduce the acquired charge. Particles of irregular shape would require auxiliary calibration to determine their drag coefficients.

These measurements would be performed for various particle sizes, temperatures, pressures, relative humidities, and purities (and therefore electrical conductivities) of ice and water. Observations would be made to related processes such as droplet shattering and resulting charging. The ice surface roughness should also be varied. Freezing droplets onto the ice surface could be used to vary surface textures.

### D. 5. 2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-ice	4
Type	5
Pollutant	
Pressure	
Temperature	4
Relative humidity	4
Charge	3
Rate of cooling	
Time	
Sound	3
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	4
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	



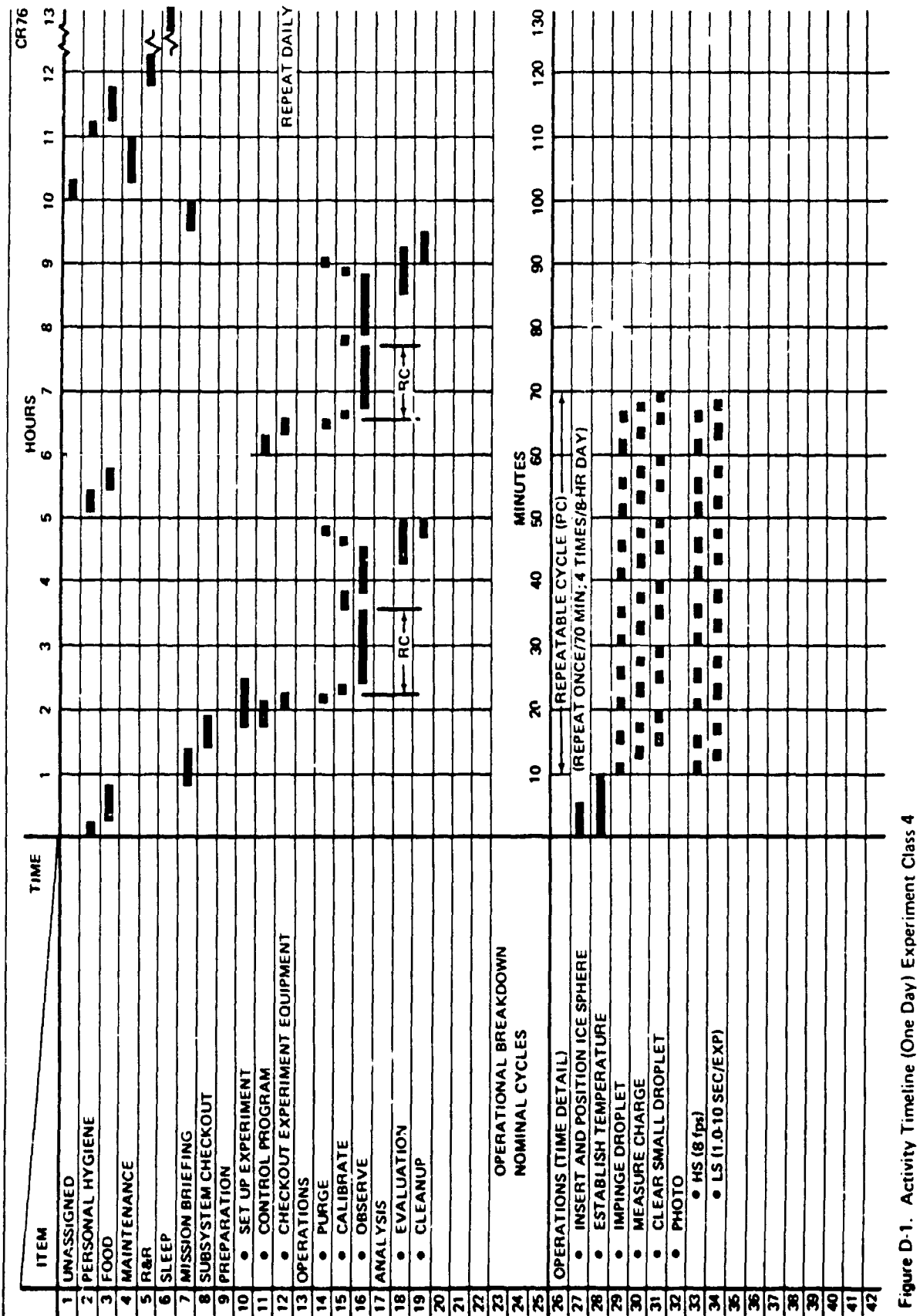
## D.6 PROCEDURE

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with thought to translating this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient (see Figure D-1).

A static diffusion ice chamber will be utilized for these experiments. Optical windows are required on the sides for visual and photographic observations. The upper plate temperature ranges from  $+10^{\circ}\text{C}$  to  $-40^{\circ}\text{C}$  and the lower plate from  $-10^{\circ}\text{C}$  to  $-40^{\circ}\text{C}$ .

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Purge chamber</li> <li>• Establish thermal and vapor equilibrium</li> <li>• Photograph (time lapse)</li> <li>• Insert pellet and position</li> <li>• Apply static polarizing electric field</li> <li>• Impinge droplet</li> <li>• Apply appropriate charge measuring electric field (resulting motion being photographed)</li> <li>• Turn off charge measuring fields</li> <li>• Recycle with other droplets (50 events) (purge when necessary)</li> <li>• Recycle with other droplet diameters (4 sizes)</li> <li>• Recycle with other humidity levels (4 levels)</li> <li>• Recycle with other temperatures (4 values)</li> <li>• Recycle with other impurities in ice and water</li> </ul>	<p>5</p> <p>10</p> <p>5</p> <p>2</p> <p>3</p>



Appendix E  
CLASS 5  
ICE CRYSTAL GROWTH HABITS

E.1 ICE CRYSTAL GROWTH HABITS APPROACH 1

E.1.1 Introduction

After a comprehensive study of the ice crystals collected in clouds Weickmann (1947) observed that the type or habit of ice crystals in clouds are determined by the cloud temperature. The strong correlation between crystal habit and temperature were later confirmed by many observers for ice crystals in clouds and in laboratory cloud chambers. Notable experiments in the laboratory were performed by Aufm Kampe et al. (1951), Mason (1953), Hallett and Mason (1958) Kobayashi (1961). These workers investigated the effect of supersaturation and temperature of ice crystal habit and found that the habit is strongly determined by the temperature and observed the existence of secondary features such as elongated crystals at or above water saturation at specific temperatures. For example at -5C one gets needle shaped crystals and at -15 C dendritic crystals.

A relationship between the existence of these elongated shapes and the growth rate is evident from the growth rate experiments of ice crystal both from laboratory and field experiments (for example, Hallett 1965, Isono et al 1956). A theoretical study on the growth rate by Jayaweera (1971) indicated that after a growth time of 7 minutes the growth rate is dominated by the crystal shape. A reasonable agreement between the model prediction and laboratory experiment was shown recently by Ryan et al. (1974 to be published).

All experiments so far performed suggest a close relation between the crystal habit and growth rate. At the same time both these properties are highly dominated by temperature. But so far no mechanism for the

dependence of temperature has been put forward. The growth rate of ice crystals is the controlling factor for the rate of conversion of liquid water to ice crystals, hence determines the efficiency of the Bergeron - Findeisen mechanism. In modelling the ice crystal process, one has to know the growth rates, which in turn depends on the crystal habit. Although our present knowledge suggests a dependence of crystal habit on temperature as far as their basic habits are concerned, the conditions for secondary features such as needles and dendrites are complicated by their dependence of supersaturation. The evidence for the importance of cloud environment on crystal habit is shown further by Jayaweera and Ohtake (1974). It is important therefore to study the growth rate and habit for various temperatures and supersaturation so that a clear understanding of the ice crystal properties that occur in various clouds and at different sections of the same cloud can be better understood.

#### E. 1.2 Objective

Determine the temperature, pressure and relative humidity conditions which dictate ice crystal geometry and growth rate under pure diffusion (non-convective) condition.

#### E. 1.3 Scientific Justification

Ice crystals are responsible for releasing much of the precipitation (snow, rain, hail) from clouds in the high- and mid-latitudes. The growth properties of ice crystals form an important aspect in the area of weather modification. The cold precipitation mechanism depends on the conversion of water vapor into ice crystals. The rate of this conversion is controlled by the growth rate of ice crystals, which in turn is governed by the distribution of water vapor and temperature around the crystals.

Needle and dendritic ice crystals have been found to grow very rapidly at temperatures of  $-3^{\circ}\text{C}$  to  $-5^{\circ}\text{C}$  and  $-12^{\circ}$  to  $-16^{\circ}\text{C}$ , respectively. At other temperatures, the linear growth rates are much slower.

The growth rate determines the rate of release of latent heat during the freezing process. This will effect the bouyancy of the cloud, and therefore the cloud

dynamics. It is well established that cloud dynamics plays a important role in the release of precipitation. Certain crystal types enhance collision and riming processes while the potential breakup of crystals becomes important in the cold precipitation process of conversion of super-cooled water droplets to ice. Ice breakup may also play an important role in thunderstorm electrification. Ice crystal growth rates and their controls need to be better understood before this knowledge can be efficiently utilized in the "when and where" decisions of weather modification. These are only a few of the many areas where detailed knowledge about crystal growth habits are needed.

#### E. 1. 4 Applications

- 1) Weather modification
- 2) Numerical cloud modelling
- 3) Thunderstorm models and forecast
- 4) Physics of ice

#### E. 1. 5 Terrestrial Laboratory Limitations

The interesting enhancements in the growth rate, and peculiar growth habits of ice crystals occur at relatively long times of growth (few minutes).

As such the present day researchers on ice crystal habit and growth rates are restricted to make these studies in the field or in large laboratory cloud chambers. Field experiments although allow for the required long time of growth have no control of the environmental conditions. On the other hand laboratory cloud chambers restricts the growth times and if the chambers are made bigger to allow for longer times of growth then the uniformity of the environment has to be penalized. Either way the terrestrial experimenters are limited by the restrictions of either not having enough time or non uniform environmental conditions or both. Use of suspension devices interfere with the heat transfer and local disturbances and anomalies in vapor pressure and temperature may result.

Present laboratory investigations of the pure diffusional mechanisms involved in the crystal growth are hampered by gravity-induced convection currents and restricted by the need of mechanical supports. These restrictions

modify the heat and vapor transport characteristics sufficiently to mask the observation of the desired physical processes. Ventilation is important in the growth of ice crystals and the contribution of this factor can be better evaluated by comparing terrestrial wind tunnel data with the low-gravity diffusional growth data.

Past and present terrestrial laboratory experiments have shown the complexity of the pre-precipitation ice growth phase within a cloud. Important questions remain as to just what critical parameters of pressure, temperature, and vapor concentrations determine the transitions between crystal growth types.

A low or zero-gravity platform would greatly enhance the study of ice crystal growth habits. Gravity-induced convection would be reduced by a factor related to the reduction of the residual acceleration (gravity and vehicle motion) and at the same time permit extended periods of time for crystal growth without physical supports.

#### E. 1.6 Quantification

Achievements of the critical parameters of pressure, temperature and relative humidity on the pure diffusional growth rate and habit of ice crystals by terrestrial experiments will also be hindered by the inherent difficulties mentioned above. At present our knowledge is sufficiently advanced in spite of these restrictions to relate various properties of ice crystals with each other but still insufficient to understand the mechanism of the formation or growth of ice crystals. Advances may be anticipated in theoretical studies but these verifications will not be possible from terrestrial experiments. It is hoped that zero gravity experiments will provide the necessary information so that theoretical models may be verified against them. Proper understanding of the growth mechanism will only be achieved through an interplay of theoretical modelling followed by experiment verification.

### E. 1.7 Approach

#### E. 1.7.1 General

The growth rates of ice crystals are a sensitive function of the relative humidity and the growth habit also depends on the humidity if the environment is below water saturation. Therefore the primary requirement for this experiment is the accurate measurement and stringent control of temperature and humidity. An experiment designed to study the growth rate of ice crystals, would require a large number of repetitive experiments of growing an ice crystal to a desired time and melting it to determine the mass. Alternately one can use large numbers of ice crystals and grow them together and melt them at successive intervals to determine mass at various time intervals. However this latter method will interfere with the humidity of the temperature of the chamber. Therefore a compensation is to use a diffusion chamber and place ice crystals so that each crystal is subjected to the specific temperature of the environment and allowed to grow for a known time and melting all crystals and determine the mass.

#### E. 1.7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	4
Type	6
Pollutant	
Pressure	3
Temperature	4
Relative humidity	4
Charge	
Rate of cooling	
Time	
Sound	3
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	



<u>Parameters</u>	<u>Variations</u>
Optical	4
Shape	4
Orientation	3
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### E. 1. 8 Procedure

General activity details are given below followed by a representative activity timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient. (See Figure E-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Unassigned</li> <li>• Personal hygiene</li> <li>• Food</li> <li>• Maintenance</li> <li>• R &amp; R</li> <li>• Sleep</li> <li>• Mission briefing</li> <li>• Subsystem checkout</li> <li>• Preparation</li> </ul>	12 18 30 48 48 480 30 30
Operations Breakdown	
<ul style="list-style-type: none"> <li>• Operation (time detail)</li> <li>• Purge</li> <li>• Establish temp between plates</li> <li>• Establish pressure of chamber</li> <li>• Measure temperature at plates</li> <li>• Measure RH at plates</li> <li>• Measure pressure of chamber</li> <li>• Insert ice crystal or ice nuclei</li> <li>• Allow crystals to grow</li> <li>• Obtain time lapse photograph</li> <li>• Measure crystal temperature</li> <li>• Melt ice crystals</li> <li>• Photograph</li> <li>• Data dump</li> </ul>	5 5 0.5 0.5 0.5 2 10-100 10-100 5 1. 0.5 0.5

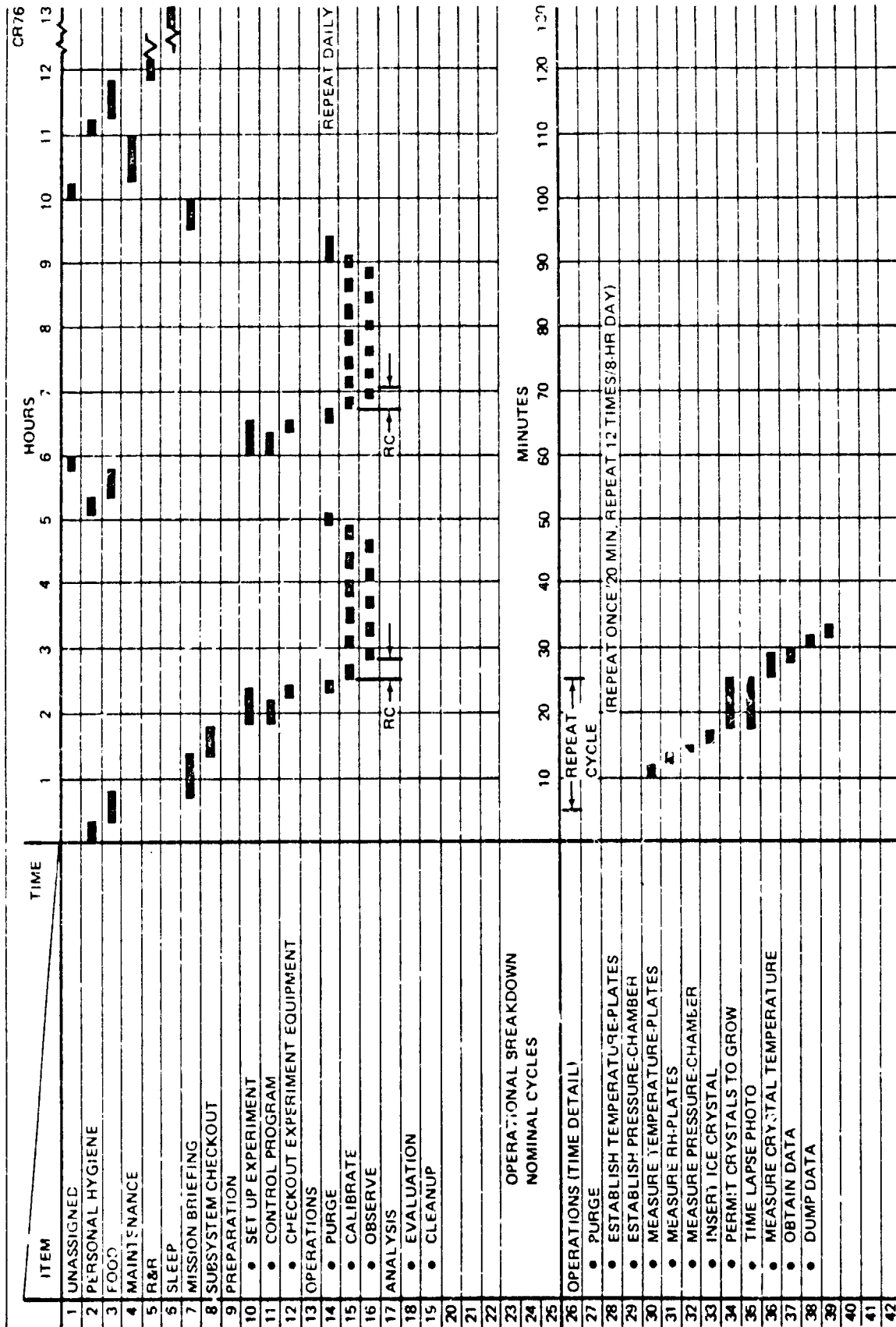


Figure E-1. Activity Timeline (One Day) Experiment Class 5

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## E.2 ICE CRYSTAL GROWTH HABITS APPROACH 2

### E.2.1 Introduction

Previous work on habit of ice crystals growing from vapor show:

- (1) influence of temperature on habit
- (2) influence of supersaturation on habit (see Table E-1).

A static diffusion chamber is employed to obtain data on ice crystal growth habits. An ice plate thermal diffusion chamber is used to investigate low supersaturations (Hallett and Mason, 1958). Growth rates are influenced by the thermodynamic properties of the carrier gas: thermal conductivity and vapor diffusivity. The theory is discussed for transfer from a freezing water drop by Johnson and Hallett (1968) where the growth rate is shown to be proportional to  $K_g + \beta D L_v$

$K_g$  = gas thermal conductivity

$\beta$  = coefficient of Clausius Chapyeron Equation

$D$  = diffusivity of water vapor in gas

$L_v$  = latent heat of evaporation of ice

Growth rates have been measured in air (Hallett and Mason, 1958), in  $\text{CO}_2$  and  $\text{H}_2$  by Isono et al, 1957; Isono, 1958; Van den Heuvel and Mason, 1959; similarly, low pressure influences.

These coefficients are shown in Table E-2.

Both crystal habits and growth rates can be drastically changed by trace impurities, particularly of polar organic compounds. For example, concentrations of camphor as low as  $10^{-3}$  mb pressure can change all growth habits to needles or prisms (Hallett, 1968); a similar change may be caused by a silicone grease impurity.

In addition to these influences on growth, the motion of gas in the neighborhood of growing crystals may lead to enhanced growth rate. This was discussed by Mason (1953) for ice crystals falling at terminal velocities. A transition from plate to dendrite growth occurs at a critical size  $\sim 200\mu\text{m}$ . Studies under forced ventilation have been made by Gamara and Hallett

Table E-1  
VARIATION OF ICE CRYSTAL HABIT IN GROWTH  
FROM THE VAPOUR IN AIR

Temperature interval °C	Supersaturation				
		up to 5%	5 to 15%	15 to 100%	greater than 100%
0	-2	Plates	Plates	Dendrites	Dendrites
-2	-3			Plates	Plates
-3	-4	Solid prisms	Hollow prisms	Hollow prisms	Hollow prisms
-4	-5		Needles	Needles	Spikes at 22° to <u>c</u> axis
-5	-8		Hollow prisms	Hollow prisms	Hollow prisms
-8	-12	Plates	Plates	Plates	Plates
-12	-16			Dendrites	Dendrites
-16	-25			Plates	Plates
-25	-40	Prisms	Hollow prisms	Hollow prisms	Hollow prisms, spikes

Prisms and needles have grown more along c than a axis directions.  
Hallett (1968)

Table E-2  
VALUES OF  $K_g + \beta DL_v \times 10^4$   
 $\text{cal cm}^{-2} \text{sec}^{-1} \text{°C}^{-1}$

gas/pressure →	1	0.1	0.01	0.001 (atm)
air	0.75	2.73	20	200
H <sub>2</sub>	4.55	11.3	71	674
CO <sub>2</sub>	0.47	1.73	17.3	170

(1972) in a dynamic diffusion chamber. These results show a growth rate dependent on  $Re^{1/2}$ .

Growing crystals may exceed the ambient temperature by several degrees by release of latent heat. This may give rise to natural convection by bouyancy as the Rayleigh Number (Ra) exceeds 650  $\rightarrow$  1000, which occurs for  $\Delta T > 1/2^\circ C$  in air.

### E. 2. 2 Objective

Hence, the question arises as to the nature of the interaction of forced convection which ventilates a falling crystal, and the natural convection caused by its temperature excess and the bouyancy of the surrounding warmer gas. When crystals are growing on a fibre in a static diffusion chamber, from which the results of Table E-1 are taken, there is always present natural convection  $\sim$  several  $cm\ sec^{-1}$  which leave some doubt concerning the precise mechanism of transition from reticular growth form—as hollow prisms or hexagonal plates - limited essentially by molecular processes at the crystal gas interface to needles or dendrites limited essentially by transfer of heat and moisture by molecular processes enhanced by forced or natural convection. Experiment are now being performed in the laboratory to study forced ventilation and Zero g will permit the study of zero ventilation. Studies with electric field, growth under pressure, and with  $D_2O$  will shed further light on these processes.

### E. 2. 3 Scientific Justification

The proposed experiment will produce data on the interaction between molecular and advective transfer processes which occur during ice crystal growth. Results will be applicable to a host of complex crystal growth problems. In particular, growth of crystals in the atmosphere may be interpreted in terms of environmental parameters—temperature, supersaturation—for a given crystal size. This will enable us to infer from crystal size, habit and other surface detail, something about the nucleation mechanism of crystals caught from aircraft or at the earth's surface. Further, the extreme habits of ice crystals growing in the atmosphere (Hallett, 1965) may be subject to interaction between molecular and advection processes.

#### E. 2. 4 Applications

- (i) interpretation of growth parameters of natural ice crystals from shape.
- (ii) Deduction of atmospheric nucleation process from crystal forms.
- (iii) Industrial growth processes where forced and natural convection process overlap.
- (iv) Better understanding of molecular kinetics of crystal growth.

#### E. 2. 5 Terrestrial Laboratory Limitations

It is impossible to remove natural convection in a ground based system. It can be reduced by choice of carrier gas, but not removed sufficiently for an appraisal of growth with only molecular heat and vapor transfer.

#### E. 2. 6 Zero-Gravity Opportunities

The low gravity environment reduces the bouyancy by  $\sim 10^5$  which is equivalent to reducing temperature by  $\sim 10^{-4}^\circ\text{C}$ . This is impossible in any earth based laboratory

#### E. 2. 7 Quantification

The type of study is not possible in an earth based system, and Zero g gives a unique capability for conducting cloud physics tests.

#### E. 2. 8 Approach

Previous work has been described in Hallett and Mason (1958); Gamara and Hallett (1972). The first experiment is a habit study in a static diffusion chamber, where natural convection could not be suppressed. The second study is in a forced convection system where natural convection is small compared with the wind tunnel velocity; this situation occurs with crystals falling at terminal velocity in the atmosphere, for crystals with size in excess of 1 mm diameter. Problems in these systems arose from maintaining chambers ice free for appropriate viewing, maintaining constant temperature over periods of  $\sim 10$  hours. Chamber design has passed through several phases and laboratory models are now approaching good working systems. The temperature of the top plate varies from  $+30$  to  $-10^\circ\text{C} \pm 0.1^\circ\text{C}$ ; base plates from  $-10$  to  $-50^\circ\text{C} \pm 0.2^\circ\text{C}$  with the temperatures changes taking not greater than 1/2 hour. The proposed system for low g must provide this



range of temperature. A double wall chamber is essential, with the control of the conductivity being performed by varying the gas pressure between the walls. Some deicing capability may be necessary on the inside wall and may be achieved by providing a ducting resistance strip on the inside of the chamber. Data will be recorded by 2 time lapse movie cameras, 16 mm, at 10 second to 50 second intervals. One wide angle lens (55mm) one telephoto lens (250 mm) will be used. Illumination will be from a 50 watt strip lamp with cylindrical lens. The chamber will be capable of being evacuated to as low as 1 torr. Analog recording of the top and bottom plate temperature are required and a temperature profile sensor will be used in the chamber.

#### Chamber

Base and top - brass with cooling systems.

Walls - double thickness glass (quartz) sealed unit with optical flats on each side 2 cm x 10 cm.

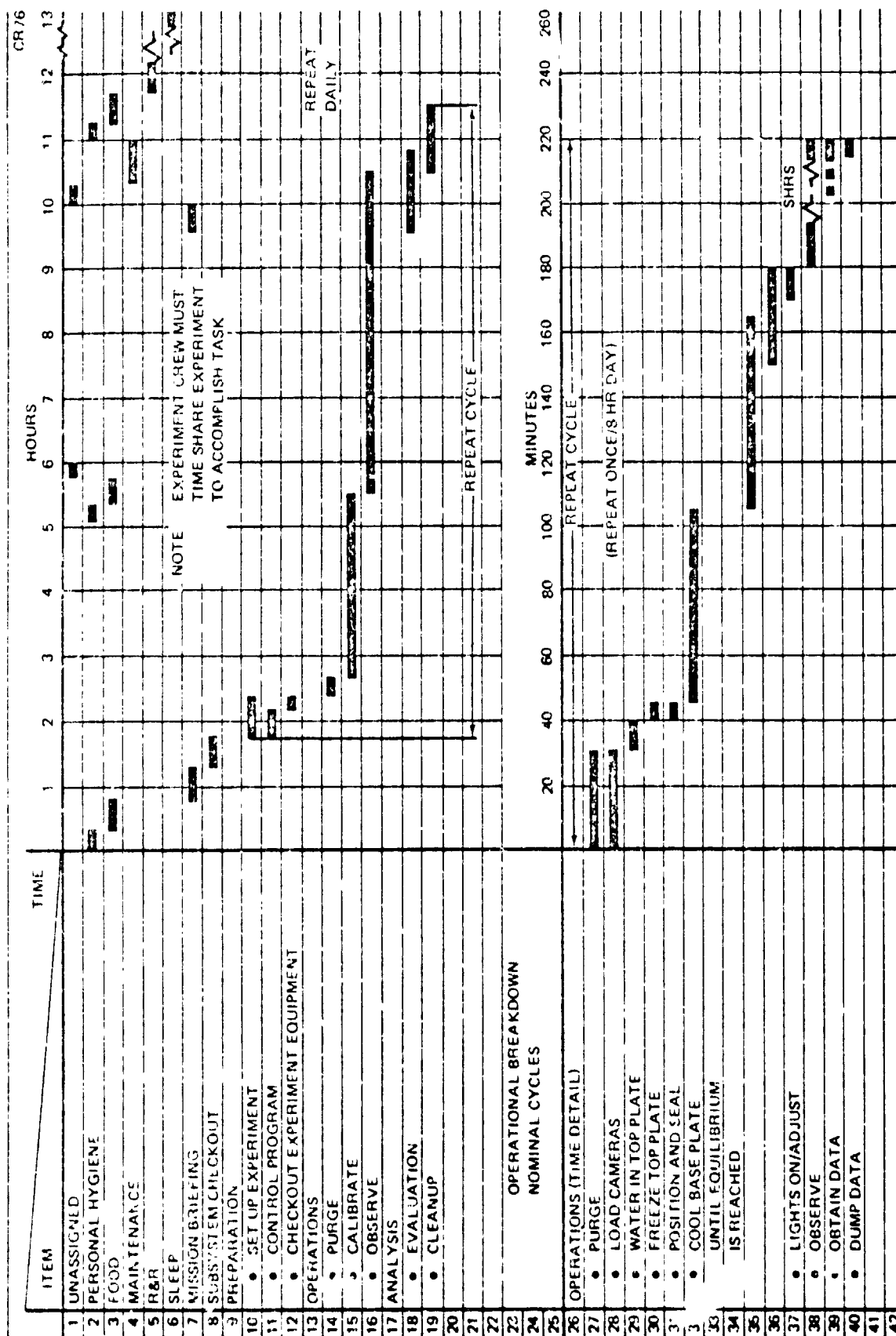
Conductivity controlled by varying air pressure between walls, monitor by Parani/gauge.

#### E.2.9 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to the low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure E-2).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Load cameras	10
● Load water into upper plate or load water into upper plate and freeze and place in position.	30
● Cool upper and lower plates to specified temperature	60
● Rain out nuclei	30
● Insert fiber	10(sec)
● Grow crystals and take movies in time lapse: high supersaturation or medium supersaturation or low supersaturation	1(hr) or 3(hrs) or 5(hrs)
● Warm up chamber, melt crystals, clean out for next run	60



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Appendix F  
CLASS 6  
SCAVENGING

F.1 SCAVENGING APPROACH 1

F.1.1 Introduction

The scavenging of nuclei and particulates by cloud droplets is a transport phenomenon that is particularly relevant in cloud physics. The scavenging process might be divided into three major divisions: (1) transport of particulates to the scavenging droplet by Brownian motion, charge effects, etc., (2) in cloud scavenging by phoretic forces (rainout) due to droplet growth, and (3) below-cloud scavenging (washout).

F.1.2 Objective

It is the objective of this experiment to investigate significant problems of cloud scavenging. The processes that go on in clouds are very complicated. There are perhaps a half dozen different mechanisms that contribute to scavenging (phoretic forces, nucleation, Brownian motion, inertial and electrical effects). So far we have not isolated these different mechanisms to determine each of their influences. From this investigation on scavenging of submicron aerosols we should obtain the numbers useful for prediction and the models necessary to predict in cloud scavenging resulting from phoretic and Brownian motion effects.

F.1.3 Scientific Justification

Scavenging of particulates and absorption of gases by hydrometeors or by large aerosol particles is important because this is one of the most significant and inexpensive way of cleaning the air. However, the complexity of problems and number of parameters (which must be included in any successful model) means that a relatively small number of scavenging models are applicable for wider size spectrum of particulates in the atmosphere. One of the major problems is related to the interaction between the hydrodynamic

forces, Brownian motion and forces effective mainly on a distance comparable with the free path of air molecules (different kinds of phoretic forces, London forces, etc.).

There is strong evidence that many ions were bound most intensely on smallest fog or cloud drops (J. Podzimek, et al., 1956) which had a very small settling velocity. Also in question are the mechanisms which cause the passivation of giant hygroscopic nuclei by non-active tiny aerosol particles or by surfactants. For these reasons, it is very important to know, for example, how the phoretic forces will influence the binding of tiny aerosol particles or gases on the surface of cloud or precipitation elements in the absence of a gravitational field. A similar approach to the solution of the problem offers better comparison of the experimental data with specific types of solutions to the Fokker-Planck equation or other equations describing the diffusional and phoretic flux onto the cloud elements and giant nuclei.

Stefan (1887) reported on a "dust-free zone" around an evaporating body and Tyndal (1870) observed that aerosol particles move toward the body having lower temperature. Facy (1955) described the particle-free space around an evaporating drop and the accumulation of aerosol particles on a drop growing by condensation of water vapor. The studies of the influence of thermophoresis and diffusiophoresis on the binding of particles was undertaken by several authors: Rosenblatt and LaMer (1946), Saxton and Ranz (1952), Schnitt (1959), Schadt and Cadle (1961), and Prokhorov and Leonov (1961). Most of the authors used aerosol particles (oil drops) suspended in a Millikan condenser. Schmidt and Waldmann, using mixtures of different gases like  $N_2$ ,  $H_2$ ,  $C_2H_2$ ,  $O_2$ , Ar,  $CO_2$ ,  $C_3H_8$ , and  $C_2H_6$ , found good agreement with the theory that the oil droplets were moving in the direction of the flux of heavier gas molecules. The experiments of Prokhorov and Leonov, (1961) investigating diffusiophoretic forces using a special torsion balance, were to a certain degree inconclusive. Goldsmith et al. (1961) investigated the influence of Stefan flow on nichrome particles tagged with thorium B and moving in a narrow channel with one wall wetted by water and the other covered by a highly water-absorbing surface (sulfuric acid). They were able to prove the general validity of the Stefan's flow velocity toward the wall on which

condensation of water vapor occurs. However, they found large discrepancies between the theoretically calculated influence of the thermophoresis and their experimental results that was probably due to the theoretical complexity of the case of a larger particle ( $d > 0.1 \mu\text{m}$ ) moving under the influence of thermophoretic force.

The theoretical approach to the description of the behavior of an aerosol particle in a given temperature or gas (vapor) density gradient can be divided into three special cases according to the size of aerosol particles.

For a particle much smaller than the molecular mean free path ( $\text{Kn} > 1$ ), Deriaguin, Kukhin, et al. (1957, 1959) and, independently, Schmitt (1959) deduced for the thermophoretic velocity a formula very similar to the formula obtained by Einstein in 1924. For diffusiophoretic forces, Deriaguin and Dukhin (1956) constructed an oversimplified hydrodynamic model, which was later improved by Deriaguin and Bakanov (1957). A more convincing approach is that of Waldmann (1959), based on the momentum transfer method with the assumption of an isothermal gas mixture under steady state conditions. The simplified formula for Stefan flow velocity in accordance with Waldmann's model was used by Podzimek (1962) and by Goldsmith et al. (1963) in modelling the aerosol scavenging process inside clouds and human lungs.

The case of the movement of a large aerosol particle ( $\text{Kn} \ll 1.0$ ) in the temperature field was treated by Epstein (1929) who solved the simplified Navier-Stokes equation taking into account the heat conduction both in the thermally stratified medium and inside of the particle. The most important result of Epstein's calculation was the fact that the thermophoretic velocity does not depend on the size of the particle. For higher heat conductivity, as found in a majority of solid aerosols, we have to use more of the sophisticated models of Brock (1962) or Deriaguin and Bakanov (1962). Both authors tried to take into account the influence of thermal slip, which results from the coupling between the temperature and velocity fields, and of the temperature jump and the friction slip at the particle surface. Deriaguin and Bakanov (1962) and Deriaguin and Yalamov (1964) presented more sophisticated models, the first of which unfortunately does not contain the dependence on the particle size. Schmitt and Waldmann (1961) deduced an approximate

formula for larger aerosol particles in a diffusion field taking into account the force of a diffusion slip on a particle moving according to the Stokes resistance law. In order to obtain better agreement with experiment, they proposed that a correction factor be incorporated into their equations. Another correction term was introduced by Brock (1963) in his study of this problem. Deriaguin et al. (1961) derived independently a simplified expression for the diffusiophoretic velocity of a particle of nonvolatile substance moving with the center of gravity of the air-water vapor system and discussed its application to the retention of aerosol particles in human lungs.

In addition there are a few papers covering a wider range of sizes of aerosol particles that utilize a realistic model based on non-equilibrium gas theory (Chapman-Enskog model). Some more general assumptions using the Chapman-Enskog model for inelastic collisions were made by Annis et al. (1973) who presented the results of a theoretical study of the motion of a spherical aerosol particle in a diffusing gas mixture covering the free-molecule to the continuum regime. In this study fairly good agreement was obtained using Waldmann-Schmitt's theory for free-molecule and continuum regime as the two extreme cases. Of course, the model is limited to low Reynolds numbers and does not contain any terms characterizing the interaction between successive molecular collisions. This problem and others related to the physico-chemical changes of a particle moving in a phoretic field remain unsolved.

There are several other fields (such as photophoresis, London forces, or different kinds of electrostatic forces) which, in our opinion, could be much better investigated in the case of larger aerosol particles in zero-gravity conditions at different pressures and temperatures. The application of similar studies in cloud physics and in environmental problems is widely discussed in papers by Hagen (1967), Slinn (1968), Saad and Jackson (1969) and by Slinn and Hales (1971).

#### F.1.4 Applications

The applications of cloud droplet scavenging are numerous. In weather control and modification some hard numbers are needed for scavenging of  $A_g I$



by supercooled water drops, because of the important role  $A_g I$  plays in many cloud seeding attempts. Further applications of scavenging are the natural cleansing of the atmosphere by cloud elements and the use of sprays in industrial (Venturi Scrubbers) emission controls. The phoretic forces are also important in the retention of submicron particulates in the human lungs.

#### F.1.5 Terrestrial Laboratory Limitations

In a gravitational environment the time allowed to study scavenging effects of particulates by cloud drops is limited by their settling velocity. In clouds however the updraft allows particulates and the cloud elements to interact over longer periods of time, and so in terrestrial labs the droplet has to be supported or the scavenging times must be short.

#### F.1.6 Zero-Gravity Opportunities

In a near zero-gravity environment the influence of scavenging over longer periods of times can be studied, and of course in a zero-g lab there would be no settling velocity to mask the effects of phoretic forces. The near zero g-gravity lab thus provides the best environment for investigating the effects of scavenging over longer periods of time.

#### F.1.7 Quantification

Knowing the scavenging efficiencies of  $A_g I$  nuclei in a cloud environment would give insights into different techniques of cloud seeding that could yield better results in weather control and modification stemming from cloud seeding.

#### F.1.8 Approach

##### F.1.8.1 General

In using a fast expansion chamber to study atmospheric processes, such as nucleation, activation, growth, etc., it quickly becomes obvious that gravity-induced convection as well as heat influx from the walls places a severe restriction on the observation times available. To offset the short observation time, the chambers are normally operated at supersaturations far greater than those occurring in the atmosphere in order to accelerate droplet growth times so that the whole experiment can be accomplished in the available time.

While this permits the time scale of many processes to be compressed to the required duration, it also makes it extremely difficult to relate laboratory observations to events occurring in the natural atmosphere with any degree of certainty.

The obvious solution is to extend the time during which useful observations can be made in the chamber. This can be attempted in either of two ways: by increasing the size of the chamber to reduce the surface to volume ratio and remove the walls far from the region of interest, or by cooling the walls at the same rate as the gas is cooled by expansion. The first method, being utilized in the U.S.S.R., involves enormous chambers which are expensive to both construct and operate and involves many complex engineering problems such as sample production, cleanliness, control, and data acquisition, due to the size. The second method can be used for chambers of reasonable and convenient size and was selected for use in building the chamber at the University of Missouri-Rolla (UMR).

The UMR group spent two years testing various freon refrigeration systems for controlling the temperature of the chamber walls before deciding that this type of system did not have either the degree of temperature uniformity and response required or the flexibility of automatic control which was desired. Attention was then turned to the use of thermoelectric modules. This type of system, in addition to satisfying the requirement for uniformity of cooling, has an additional advantage that total electrical control can be achieved. After additional testing, thermoelectric cooling was selected for use in the University of Missouri-Rolla chamber. Our four years of experience and evaluation indicate that this is by far the best way to go for the zero-g chamber.

The chamber design was then finalized based on a ten-sided prism with a sensitive volume approximately 18 inches in diameter and 24 inches tall, giving a volume of about  $3.5 \text{ ft}^3$ . The walls consist of thermoelectric cooling modules clamped between an inner wall of  $1/4$  inch black anodized aluminum plates and an outer 3 inch thick aluminum heat sink. The choice of a segmented design for the inner wall was based on the thermoelectric mounting

techniques in use at the time the chamber design was finalized. New mounting techniques have made this unnecessary and new chambers can be designed with a minimum of joints in the inner wall. This also makes an easy job of sealing the chamber, which has been the only real problem we have encountered using the older design.

As the size of the system and the number and complexity of the control and data acquisition functions became evident, it was determined that an analog control and data acquisition system would not have the required accuracy and flexibility. The decision was then made to use a digital system build around a NOVA 800 minicomputer. While this system is not yet operational, the decision not to use an analog system was strengthened by our experience with a temporary analog control system built to carry out the initial thermodynamic tests of the chamber. While the analog system does work, it has an extremely limited range, less accuracy than desired, and must be continually "tweaked up" to maintain maximum accuracy. State of the art techniques were employed so we feel we effectively reached the technological limit of accuracy.

The large size of the system and the planned use of a flow-through purge technique dictated our decision to use a continuous flow filter/drier system in place of tank air. This is coupled with a flowing water humidifier to provide clean air having a known vapor content required for sample preparation. This system as designed is not feasible for use in a zero-gravity environment. The smaller size of the proposed zero-gravity chamber does permit use of tank air and humidification can be accomplished using a porous ceramic surface to which water is supplied by capillary action. Such techniques have been successfully employed before.

Thermal control throughout the system, with the exception of the inner wall of the expansion chamber, is maintained by numerous thermally regulated circulator baths and closed loop liquid heat exchange circuits. Due to the inefficiency and large mass of this type of system, alternate models are strongly recommended for use with a zero-gravity chamber. Transistor thermometers developed at UMR are used throughout the system for temperature measurement with excellent performance.

Pressure measurements within the expansion chamber are made using a 1 psid pressure transducer referenced to a dead weight standard. While this method can not be adapted to a zero-gravity environment, the use of an absolute pressure transducer is feasible; however, some loss of resolution will occur. Pressure changes are accomplished by using an external piston driven by a precision stepping motor through a micrometer screw. Piston motion is controlled by the computer which compares the chamber wall temperature to the gas temperature, calculated from the pressure and corrected for latent heat release by the cloud, and causes the piston to move either in or out to give a zero temperature difference.

Several aerosol generation techniques have been considered, including furnaces, evaporation/condensation, bubbling of air through a solution, desiccation of a solution spray, and gas phase reactions. At present we are planning to use the desiccation of a salt spray for the first experiments. Characterization of the aerosol includes: electromicrographs, Knollenberg counter, multiangle laser scattering in a separate chamber, and activation supersaturation measurement using continuous flow thermal diffusion chambers.

Real time measurements of the liquid water content, drop size distribution, and drop concentration have proved to be a difficult problem. Initial work using laser scattering techniques were based on the measurement of the disymmetry ratio of light scattered at two angles symmetric about  $90^\circ$  and the attenuation of the unscattered beam. This method has been developed to the point where we believe it can measure the mean drop size and total liquid water content provided the droplet size distribution is not too broad. The technique does require additional refinement and calibration but has the advantage of not being gravity-dependent. This technique is not capable of providing information about the droplet size distribution.

For use with the UMR chamber, we have abandoned the disymmetry ratio technique in favor of a Doppler shift technique proposed and developed by Dr. Jerry Gollub of Haverford College. This approach uses the Doppler shifted component of scattered laser light to measure the fall velocity of the

droplets. The drop sizes are related to the fall velocities by Stokes' Law. The concentration of drop in each size range is determined from the amplitude of the corresponding frequency component of the scattered light corrected by the Mie theory function. Once the concentration and size distribution have been determined, calculation of the liquid water content is a simple matter. While this technique is excellent in a terrestrial laboratory, its dependence on the gravitational settling of the drops makes it unsuitable for use in a zero-gravity environment.

Experiments on scavenging of Aitken nuclei by cloud droplets will require the use of two separate nuclei generators. The first will be used to produce giant condensation nuclei which will serve to create a cloud in the expansion chamber. The second nuclei generator will produce the Aitken nuclei to be scavenged by the drops.

After generation the two sizes of nuclei are mixed in an appropriate concentration ratio and the concentration of giant nuclei adjusted to yield the desired drop concentration in the expansion chamber. The resulting bimodal nuclei sample is introduced into the expansion chamber at the predetermined initial pressure, temperature and relative humidity. After the chamber has come to thermal equilibrium an adiabatic expansion is used to create a cloud of drops on the giant condensation nuclei. This cloud is then given time to scavenge the smaller Aitken nuclei. After the desired scavenging interval has elapsed the chamber is opened to permit a sample of droplets to be drawn out and collected in an impactor.

An electron microscope slide is used for the collection surface of the impactor. The surface of the collector will be treated so that after the sample has been collected the water can be removed by storage in a desiccating environment and a replica of the drop will be left. The sample will then be examined with an electron microscope to determine the number of drops which did and did not contain a scavenged Aitken nucleus.

This technique will require that the Aitken nuclei be tagged in some manner to permit their identification during examination with the electromicroscope. One method would be the use of latex spheres as the Aitken nuclei.

#### F.1.8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	2
Type	2
Pollutant	3
Pressure	3
Temperature	3
Relative humidity	3
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

### **F.1.9 Procedure**

The detailed procedural steps and time required are defined in the following procedure table.

At the present time until a more detailed knowledge of the techniques that will be used in the final version the activity time line for the zero-g experimental procedure is the same as that developed for the terrestrial experiment (see Figure F-1).

It is possible that purge times and rates can be reduced but this can be determined only by testing of the actual equipment. The degree of automated control will effect the time required for some operations in both the zero-g and terrestrial experiments.

The primary change in the time line for zero-g operation will be an extension of the duration of the expansion/contraction cycle to make full use of the zero-g environment.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Turn on all electrical components	5
● Check operational readiness of components	5
● Check and calibrate control and measuring circuits	10
● Turn on noncondensable gas supply and sump pump	15
● Open expansion chamber ports	5
● Purge system	10
● Load control program into control computer	(5)
● Load time-lapse camera	(3)
● Close expansion chamber ports	0.5
● Turn on thermal controls: conditioning chamber, humidifier, expansion chamber	0.5
● Adjust gas flow to humidifier	0.5
● Adjust gas flow to aerosol generators	0.5
● Turn on aerosol generators	1
● Turn on hygrometer, particle mass meter, and particle size meter	1
● Monitor sample for relative humidity, particle size, mass and concentration	10
● Adjust humidifier and aerosol generators as required	(1)
● Open expansion chamber ports	0.5
● Transfer sample monitor point	0.5
● Monitor sample for relative humidity, particle size, mass, and concentration	8
● Close expansion chamber ports	0.5
● Turn on expansion mechanism, pressure control gas temperature, video, liquid water meter and drop size meter, laser	1



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Let expansion chamber come to thermal equilibrium	1
● Turn off: aerosol generators, thermal controls for conditioning chamber and humidifier, hygrometer and particle size, mass and concentration meters	(2)
● Purge system (except expansion chamber)	(4)
● Turn off noncondensable gas supply and sump pump	(1)
● Turn on time lapse camera	0.5
● Start expansion/cooling cycle	30
● Observe chamber on video monitor	(-)
● Turn on sump pump and sampling impactor	0.5
● Take impactor sample from expansion chamber	1
● Turn off sump pump and sampling impactor	0.5
● Stop time lapse camera	0.5
● Turn off pressure control, expansion mechanism, video, laser, liquid water meter, and drop size meter	1
● Heat chamber to +10°C and turn off expansion chamber thermal control	5
● Open expansion chamber ports	0.5
● Turn on noncondensable gas supply and sump pump	0.5
● Purge chamber	8
● Unload time-lapse camera	(3)
● Transfer impactor sample to storage	(5)
● Close expansion chamber ports	0.5
● Turn off noncondensable gas supply and sump pump	0.5
● Check computer data	4
● Transfer data to permanent storage	1
● Shut down system or start next experiment	10

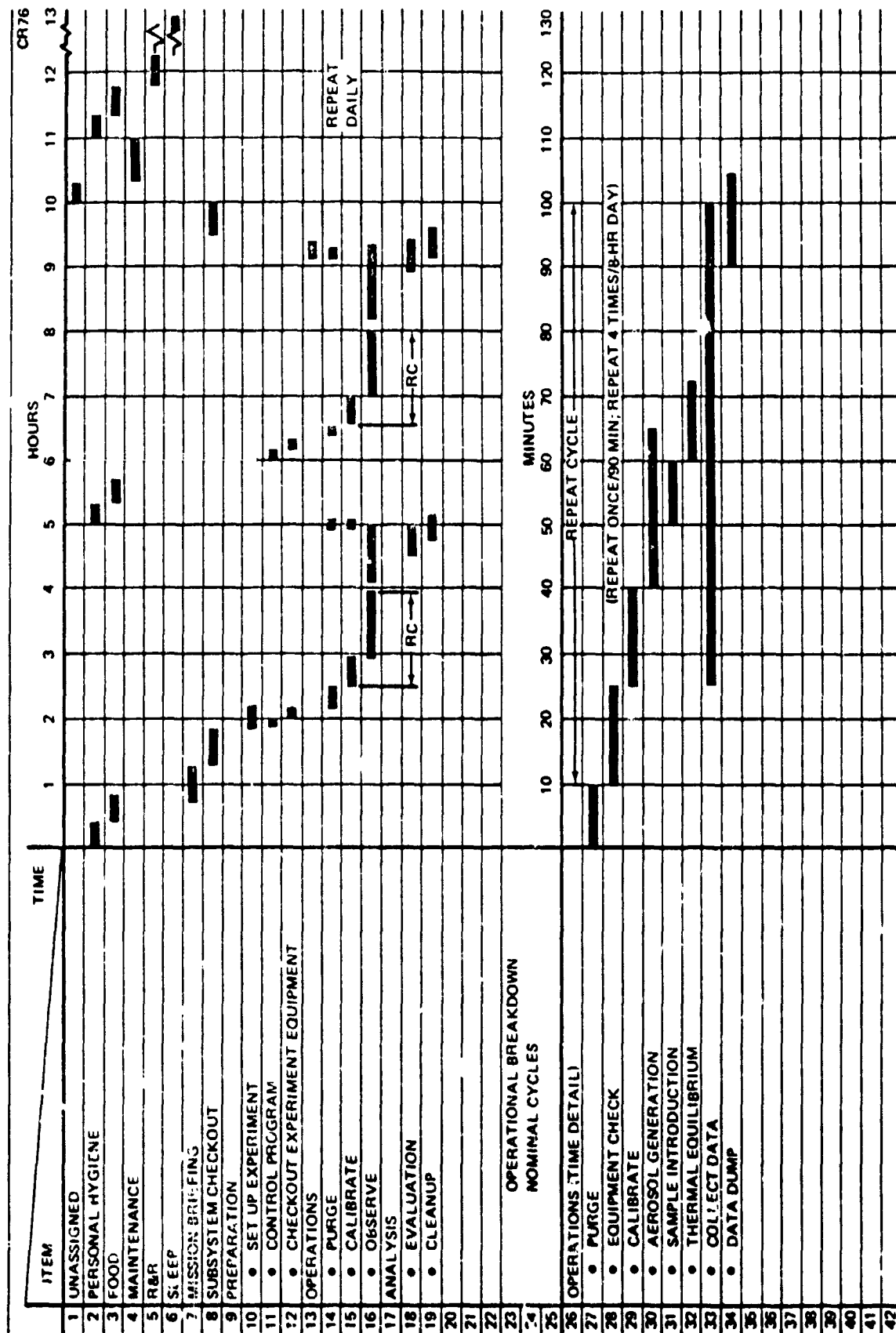


Figure F-1. Activity Timeline (One Day) Experiment Class 6

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## F.2 SCAVENGING APPROACH 2

### F.2.1 Introduction

Atmospheric scavenging is the removal of particulate or gaseous matter by hydrometeors. The importance of understanding this phenomenon applies not only to cloud physics, but also the study of air pollution. This is the reason why for years a majority of investigations in this field was sponsored by the AEC and currently also by EPA to whom scavenging is mainly a transport of foreign matter from the troposphere to the ground. Cloud physicists have become increasingly concerned with scavenging mainly in the context of nucleation: on one hand, particles might act as nuclei through a scavenging process, but on the other hand, nuclei might be removed by scavenging before they encounter the necessary conditions to nucleate.

Several mechanisms have been identified which contribute to the capture of particles by hydrometeors. To what degree each of these mechanisms is responsible for the scavenging action depends largely on the size range of the particles in question. In a general way equation (1) describes the decrease in particle number density  $n_p$  due to the capture by drops (number density  $n_d$ )

$$-\frac{dn_p}{dt} = K_{pd}(D_p, D_d, \dots) n_p n_d \quad (1)$$

whereby the collision parameter  $K_{pd}$  is strongly dependent on the particle and drop diameters  $D_p$  and  $D_d$ , but also on their densities, surface properties, etc., as well as on the carrier gas (humidity, temperature, turbulence).

$K_{pd}$  can be written as the sum of the collision parameters describing individual capture mechanisms (we assure here that collisions result in capture):

$$K_{pd} = K_b + K_{th} + K_{diff} + K_{sh} + K_s + K_e \quad (2)$$

where the subscripts b indicates Brownian motion, the thermophoresis, diff diffusiophoresis, sh shear (turbulence), s sedimentation and e electrical effects. The theoretically derived values for  $K_b$  have been verified in the area  $0.001 < D_p < 0.1 \mu m$  where a strong decrease with increasing  $D_p$  is the

main feature; for smaller particles the question arises how to interpolate for the transition to the free molecule regime. On the other hand, in the case where the drops have an appreciable fall velocity, Slinn and Hales (1) proposed a correction for ventilation (theoretical). If particles are typically larger than  $\sim 0.5 \mu\text{m}$   $K_{sh}$  and  $K_s$  increase strongly with increasing  $D_p$ . If, and under what circumstances the  $K_{pd}$  minimum in the  $D_p = D_d$  area (Greenfield gap) is bridged by  $K_{th}$  and  $K_{diff}$  or possibly  $K_e$  is quite uncertain. Most work on particle interaction due to phoretic forces has been theoretical (e.g., Slinn and Hales, 2), and the few experiments are of rather qualitative nature (e.g., Vittori and Prodi, 3).

Recent experiments were mostly carried out with scavenging droplets being  $\geq 100 \mu\text{m}$  in diameter. Vertical windtunnels as the one at UCLA, or drop tubes (Clarkson College, Potsdam, New York or Illinois State Water Survey) are the most successful tools. Considerable work with natural drops has been performed at Battelle NW (4). A unique laboratory experiment was carried out by Mack and Katz (5) whereby fog drops of 3 to 15  $\mu\text{m}$  diameter scavenged fluorescent particles of  $\sim 2 \mu\text{m}$  diameter in Calspan Corporation's 600 m<sup>3</sup> expansion chamber.

All experiments mentioned above have in common the fact that the combined effect of several collision and capture mechanisms can be measured. Depending on the particular combination of particle and drop sizes the fall velocity of the drops is more or less influential.

### F.2.2 Objective

The purpose of the experiments is the measurement of those collision parameters for drop-particle combinations which are not related to gravity induced mutual motion of drop and particle. In turn, subtracting these results from those of corresponding terrestrial measurements one obtains values for the effect of orthokinetic (sedimentation) scavenging only.

### F.2.3 Scientific Justification

There is great need for increased knowledge of the various collision parameters (or collection kernels) and collection efficiencies for a wide range of particle and drop sizes. In virtually every situation where droplets and

particles coexist it would be beneficial to better understand the mechanisms at work. A typical example is weather modification: particles are released into the atmosphere where they are assumed to perform according to a certain model; however, if the model is incorrect due to insufficient knowledge of particle-droplet interaction the result could be quite different from expected, e. g., a loss of particles due to scavenging before they can fulfill their intended role. Similarly, where wastes are released into the atmosphere in the form of particulates, assessment and prediction of the impact - be it inadvertent weather modification or removal by (poisoned) precipitation - is only possible if all facets of particle-hydrometeor interaction are known. Also, in the more and more important area of pollution abatement, techniques in the suppression of particulate effluents might benefit from a better grip on scavenging mechanisms. The use of small particles as tracers in cloud research depends heavily on the knowledge of the relative importance of the various types of interaction.

#### F. 2. 4 Applications

Most applications benefitting from increased knowledge of scavenging mechanisms were mentioned above. Ranking them according to priority is a difficult task. Among cloud physicists the weather modification application would probably attract the most interest, whereas those involved in problems of air pollution most likely would give higher priority to accurately knowing the fate of pollutant particles encountering hydrometeors.

#### F. 2. 5 Terrestrial Laboratory Limitations

In scavenging experiments the limitations due to gravity are twofold:

(1) although fall tubes, and particularly vertical windtunnels are excellent facilities where individual hydrometeors can be observed for prolonged time periods, the contribution to scavenging due to the motion of the hydrometeor relative to the air cannot be suppressed. Thus, an experimental result may be a mixture of this orthokinetic scavenging and other mechanisms, and makes a correct interpretation extremely difficult if not impossible. An example is the previously mentioned investigation by Mack and Katz who found the scavenging rate to be much higher than predicted by theory which

suggests that the orthokinetic mechanism would dominate. Unfortunately, this mode of capture could not be eliminated in a sort of control experiment which would have revealed the actual contribution of sedimentation scavenging.

(2) Terrestrial experiments involving a whole cloud of hydrometeors suffer generally from a time limit imposed by the settling speed of drops or ice crystals. In most instances scavenging is a rather slow process, the time limit is responsible for the scarcity of scavenging events per experiment, thus reducing the signal-to-noise ratio.

#### F.2.6 Zero-Gravity Opportunities

From the above section it becomes obvious that a zero-g environment could be beneficial in certain scavenging experiments:

- A. Experiments in which at least a substantial portion of scavenging is thought to be attributable to the orthokinetic process could be performed at 1-g and zero-g thus revealing the actual extent of scavenging due to the settling motion of the hydrometeors.
- B. Experiments in which one wants to investigate scavenging mechanisms other than orthokinetic, but where terrestrial fall speed of the hydrometeors limits the available time below acceptable limits, or where counteracting measures (windtunnel) make it difficult to vary a primary parameter (e. g., growth, evaporation).

#### F.2.7 Approach

##### F.2.7.1 General

In atmospheric scavenging the sizes of participating particles and hydrometeors range so broadly - three orders of magnitude - that a selection has to be made as to what size combinations should be studied and what other parameters considered. The selection should not only be influenced by what information is most desirable, but also by the degree of ease with which the information can be gained. In this latter respect the suggested choice of small cloud size droplets and submicron AgI particles appears in a very favorable light because the important problem of detecting the event of particle capture is seemingly solved in a brilliant way, i. e., by contact ice nucleation. The assumption is implied that every AgI particle captured by a drop induces its freezing; this assumption, however, is quite unfounded



except perhaps for rather low temperatures where, as Katz and Pilie (6) recently showed, most ice crystals are generated by vapor deposition on AgI particles; under such conditions where a multitude of ice crystals would intermingle with the drops, the latter would rapidly evaporate thus terminating the experiment. The instability created by the coexistence of ice and water several degrees below freezing would seem to cause a problem even if only contact nucleation would take place. The very fact, however, that for experiment class 2 the suggestion is to explore the relative roles of contact, sublimation and internal ice nucleation, indicates that contact nucleation is not a desirable method of monitoring particle capture.

The basic problem, therefore, remains the method of detecting the events of particle capture by hydrometeors. Methods employed by some previous workers are:

- A. Adam and Semonin (7): particles are bacteria, each drop is cultured and thus reveals its bacteria content.
- B. Hampl et al. (8): particles are submicron AgCl, drops are collected and bulk is analyzed chemically.
- C. Vittori and Prodi (3): particles are ZnS (size ?), stationary ice crystal is photographed in UV showing the fluorescent particles being captured.
- D. Mack and Katz (5): particles are 2  $\mu$ m diameter ZnS-CdS, cloud size drops are replicated and photographed in UV. Information consists of drop size, size and number of captured particles.
- E. Maybank and Barthakur (9): in a different type experiment they used a radioactive tracer and subsequent autoradiography of replicas.

The problems of particle and drop generation and measurement are not totally independent of the capture detection method. For instance, if the particles have to serve as tracers the choice of substances is quite limited and may be difficult to reconcile with the selection of materials suitable for generation of a certain size of particles.

The question whether to expose droplets singly one by one or as a whole cloud to the particle aerosol can be answered as follows: the "single file" approach works well in 1-g and with larger drops which fall through the particle area and are collected thereafter. But small droplets are difficult to put into and remove from the particle environment on an individual basis, especially at 0-g.

Remote sensing of particle capture would be the most desirable procedure of data collecting (c.f., contact nucleation proposal), but if not possible the choice is between collecting the drops such that one liquid water sample results, or in a way that produces replicas of individual drops. In an experiment with nearly perfectly monodisperse particles and droplets the first procedure is acceptable, although only the average number of particles per drop is known, and information on the number distribution of drops containing several particles is lost. If the drops or even the particles show a considerable width in their size distribution it is important to preserve the full information about drop size and number and size of captured particles through drop replication in order to be able to correlate it with model results.

The actual collection of droplets has to be performed such that particles are not affected substantially by the process. Impaction on a sampling surface can be discriminating enough as not to precipitate particles not incorporated in drops, thus avoiding misleading contamination of the drop samples. Among sensitive coatings for replication gelatine, due to its transparency, is best suited for the present application.

In the search for a suitable particle material one of the considerations in the selection process concerns the solubility in water. Soluble particles are relatively easy to produce in large quantities by atomizing solutions, but detection would be limited to analysis of bulk water samples, and not of individual drops; the main drawback, though, is the risk of deliquescence in the saturated cloud environment which amounts to becoming incorporated in drops without being scavenged.

The most crucial requirement is detectability of the particles present in drop replicas. For the particle size range of 1 - 2  $\mu\text{m}$  fluorescent ZnS-CdS yielded excellent results; however, if smaller particles are to be detected when inspecting the drop replicas by optical microscope, the fluorescence method probably would not succeed because of the lighting limitations. Additional research is therefore necessary. There are submicron aerosols which can be produced easily, and which could be detected electronmicroscopically by their characteristic appearance; however, it is difficult to prepare drop replicas suitable for electron microscopy, and, more important, such an evaluation would be prohibitively laborious. It should be possible to find a suitable material which can be aerosolized, and which permits small particles in the replica surface to be developed into microscopically identifiable marks by chemical means. Such schemes have been developed for soluble aerosols, and, to some extent, for AgI (Koenig, 10). Should such a procedure annihilate the drop replicas a photomicrographic record would have to be prepared first; obviously, however, no movement of the particles lodged in the replicas would be permitted.

A further very important measurement is the number density  $n_p$  of the particle aerosol taken at least at the beginning and at the end of the scavenging period (basically, this could serve to determine the number of particles lost to the drops, but this procedure is far too inaccurate). One way this measurement can be accomplished is by sampling with a membrane filter and subsequent microscopic analysis, including the above mentioned "development" for small particles. Another possibility of determining  $n_p$  would be a light scattering measurement; although this requires accurate size information. The particle size should be determined by the setting of the particle generator which has to work reproducibly enough as not to necessitate a size measurement for every run.

The terrestrial experiment (Mack and Katz), serving as a guideline, can be described as follows. In Calspan's 600  $\text{m}^3$  chamber a cloud is produced by expansion ( $n_d \approx 300 \text{ cm}^{-3}$ ,  $D_d \approx 5\text{-}10 \mu\text{m}$ ); then, fluorescent powder (U.S. Radium Helecon 2267) was dispersed pneumatically ( $n_p \approx 100 \text{ cm}^{-3}$ ,  $D_p \approx 2 \mu\text{m}$ ). Drop impaction samples on gelatine coated slides were taken

periodically (300 sec) and particle filter samples were obtained shortly after dissemination and at the end (1800 sec).

The proposed expansion chamber as well as the Nakaya chamber would basically be suitable for this experiment. However, for the Nakaya chamber (SDI) the temperature limits are rather low and gradients of temperature, vapor pressure and saturation may confuse the issue, especially when phoretic forces are in question. The main drawback of the expansion chamber (E) is the limited possibility for hooking up accessories, but it is hoped that a satisfactory solution can be worked out without impairing the basic performance of E. In both chambers drop formation can be achieved by supersaturation; by injecting condensation nuclei, drop concentration can be controlled. In order to produce large drops ( $\sim 50 \mu\text{m}$ ) a special drop generator might be necessary which would inject the desired drop cloud directly into the chamber.

#### F.2.7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	3
Type	2
Pollutant	
Pressure	
Temperature	
Relative humidity	5
Charge	
Rate of cooling	
Time	6
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	

<u>Parameters</u>	<u>Variations</u>
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### F.2.8 Procedure

A typical sequence of events during an experiment would be as follows: after bringing the chamber to its desired initial conditions (temperature, humidity) the particle aerosol would be injected (fluorescent or not yet known, sub-micron) from special generators (to be designed) which should also incorporate provisions for removal of triboelectric charges. A filter sample and scattering measurement would be followed by injection of a predetermined amount of a hygroscopic aerosol as cloud nuclei from an appropriate generator (proposed also for other experiments). An expansion would follow which is slow enough as not to activate any particles except the CN; after the expansion reaches its predetermined value, a drop sample would be taken on gelatine slides with a specially designed micro-impactor. This sample should provide a baseline for the particle content in the drops. The second drop sample would be obtained after a predetermined exposure time (scavenging time) has elapsed which is set according to the needs of the particular experimental conditions. After compression warming evaporates drops a second particle sample is taken on membrane filter and by light scattering. A final purge would ready chamber for next run.

If droplet sampling by impaction disturbs chamber conditions too much it might be necessary to sample only once per run and rely on reproducibility and instead vary scavenging period from 0 to 1000 sec.

A further problem could be the capture of particles during drop formation. An interesting result by itself it could make it necessary to either inject the droplet cloud or to inject the particle aerosol after the drops reached their starting size.

The experiments concerning scavenging during growth or evaporation of drops would be quite similar, except that slow expansion or compression would take place during the scavenging period.

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### F.3 SCAVENGING APPROACH 3

#### F.3.1 Introduction

Past work in scavenging is well documented in the 1970-AEC report by Engelman (AEC 22 Symposium Series, Precipitation Scavenging). Current work is in progress at IITRI, Chicago under the direction of Knutson dealing with the affect of snow flakes on aerosol size distribution. Perkins of Battelle, N. W. has recently studied scavenging in convective storms using cosmogenic radionuclide and other inert chemicals as tracers. The Illinois State Water Survey has used indium as a tracer on Project Metromex in the St. Louis area. This is an outgrowth of the scavenging studies by Dingle of the University of Michigan, who is now working on computer models of convective scavenging. Klaar at the British Meteorological Office, Bracknell, England is continuing the work of Sax on scavenging of submicron AgI by falling drops in the laboratory. Storebo at the University of Vilnius, Lithuania USSR has worked in the laboratory and field for many years using radioactive tracers. Research on the basic processes underlying scavenging in the atmosphere is limited, testifying to the need for more powerful tools to approach the problem. Kerker's work at Clarkson College of Technology deals with scavenging by millimeter sized drops falling through a submicron aerosol. Cooper at NCAR has recently conducted exploratory work on scavenging by a supercooled drop. A paucity of basic studies of scavenging in supercooled cloud systems is a real hinderance in the advancement of weather modification and understanding possible inadvertent weather modification effects. These problems are being indirectly addressed by some nucleation studies but a direct approach to the fundamental variables involved is missing.

#### F.3.2 Objective

The specific purpose of this experiment for cloud physics is to quantitatively define the processes involved leading to the incorporation of aerosols into cloud elements and precipitation. This can take place by condensation or ice nucleation on aerosol particles, aerodynamic capture of aerosols by drops or ice crystals, deposition of particles on cloud drops by diffusiophoresis, thermophoresis, Brownian diffusion, and electrostatic attraction.

### F.3.3 Scientific Justification

The rate of contact between aerosol particles and cloud droplets and the condensation of water vapor on aerosols controls cloud formation and the formation of precipitation. This is of vital importance to cloud seeding efforts or inadvertent seeding by air pollutants. Present modelling efforts are limited because it is not possible to define the rates of the above processes. By the same token it is difficult to optimize seeding agent properties and dispersal.

### F.3.4 Applications

Application of this knowledge will improve cloud seeding procedures, cloud processes modelling, cloud and ice nuclei detection techniques, and gives us a better understanding of how hail, snow and rain forms, of how fog can be suppressed, how aerosols are removed from the atmosphere and how to predict climatic changes.

### F.3.5 Terrestrial Laboratory Limitations

A major problem has been the impossibility of simulating the proper time domain and eliminating wall effects. It is simply not possible to maintain an array of droplets for even a few minutes without settling losses. This makes it impossible to superimpose the effect of growth or evaporation not to mention the interaction with aerosols under those conditions. The action of precipitation sized elements is even more difficult requiring some means of support in terrestrial studies. Which leaves us at present with a very meager knowledge of what happens near a large drop when it freezes. A rather considerable amount of vapor is released which could for instance have a considerable influence on the propagation of the ice phase. That is, near such a drop a rather high supersaturation develops and many new nuclei can be activated.

### F.3.6 Zero-Gravity Opportunities

At the outset it must be realized that this presents an entirely new dimension to cloud physics experimentation and this opportunity should be approached in a flexible manner. As with any new tool, such as a laser, its possibilities



can not be predicted upon its inception. Therefore, this experiment should be a growth process to fully explore the opportunities that present themselves over the years of the Shuttle program. So it is important not to lock-in on some specific experiments but the initial experiments should serve more to become familiar with the new medium. At this stage the most attractive feature of the laboratory is the possibility to suspend drops for long periods of time or manipulate their movement at will. This makes detailed photographic records possible, especially at higher magnifications where motion can be minimized. One can envision such experiments as approaching a freely suspended drop with an ice crystal or nucleus and study what happens. The growth of a drop or ice crystal can be followed accurately and its interaction with aerosols analyzed in detail. One can vary the effect of superimposed forces on the same droplet for example to simulate environmental changes encountered in the lifetime of a drop. One can form a cloud, let it evaporate use the residue aerosol for a new experiment, etc.

#### F.3.7 Quantification

It is envisioned that in a 5 year period of zero gravity experimentation fundamental questions concerning scavenging of aerosols that have been raised in the 40's when cloud seeding became a reality will finally be answered in unambiguous terms. We have a body of relative good theory on the subject now but it cannot advance further for real practical applications because fundamental physical data is lacking. Theory without experimental verification is not overly useful and begins to stagnate.

#### F.3.8 Approach

It has been suggested to use a AgI aerosol for the scavenging experiments, that is, when AgI nucleus is scavenged by a supercooled drop it will freeze and is then detected by its optical characteristics. Implicit in this plan is that AgI nucleates efficiently by contact and not by condensation-freezing. The latter process would confuse the results completely. Such an assumption is not warranted from our experience. Furthermore, this approach would eliminate scavenging tests at warmer temperatures, which are also of considerable interest to cloud physics. The approach given below is based on past experience in scavenging research at the IIT Res. Inst. and ice

nucleation work at NCAR that relates to this problem. However, no direct work on scavenging is in progress as implied in the task statement.

The proposed experiment was chosen so it will give useful data on the ground and to allow for transition into the space environment for optimum results. At this point it does not seem advisable to start out with an experiment that would only work in space. The basic chamber would be the static diffusion chamber. Transition to experiments with other chambers is anticipated as experience is gained in space and particular questions arise. The fundamental experimental approach is as follows. A relatively dense cloud will be allowed to form in the central region of the chamber. Cloud density can be controlled by providing the desired concentration of cloud nuclei. At temperature below freezing the vapor flux from the warmer plate to the colder one will be low and to form the cloud it will be easier to humidify the incoming air and expand it slightly as it enters to generate cloud droplets at once. The object is then to stabilize the cloud after injecting an aerosol sample and then manipulating the temperature gradient to let the drops evaporate or grow larger or maintain neutral conditions. On earth the drops will soon settle out and the degree of scavenging will be limited in that time. To determine the degree of scavenging the drops can be examined for the presence of particles or the change in aerosol concentration in the chamber can be monitored or both. The various aspects of this procedure will be discussed below.

First, the choice of the aerosol to be scavenged is important. It should have a well defined particle size distribution or if possible be monodisperse. It is expected that various types of aerosol generators will be available on the shuttle. Commercial monodisperse generators are available in the range above 0.1 micron producing at least several 100 particles/cc. Below this size vaporization-condensation generators can be used and the size range narrowed down by use of diffusion batteries or the particles are charged and then a desired size range is selected on the basis of electric mobility. Water insoluble materials should be used so there is no change in particle size due to condensation of water vapor at the low supersaturations of interest.

Next an accurate system for counting the concentration of the aerosol is necessary. It is envisioned that periodically small aerosol samples are removed from the chamber to monitor the aerosol decay as a measure of scavenging. For this purpose it is suggested to use a condensation nucleus counter developed at NCAR. In it an aerosol sample is added to a filtered, humidified air stream that is then cooled about 15C so condensation takes place on all aerosol particles present. These drops are allowed to grow so they can be countered one by one with an acoustic counter. Low concentrations of a few tens of drops can be detected accurately. This device can be simplified for use in a zero-gravity environment because droplet settling losses are not a problem.

To determine the number of particles scavenged directly by the droplets, it is proposed to use a special filter syringe to collect droplets only. The filter would be an electron microscope grid covered with an  $\text{Al}_2\text{O}_3$  film which has small holes allowing air to pass through it. The filter is preceded by a short tube into which suddenly a cloud sampled is pulled but not far enough to reach the filter. Some of the droplets near the filter will impact on it due to their inertia. The object of this procedure is to prevent unscavenged particles from reaching the grid. After removing the filter inlet tube, the drops can be sucked through the filter leaving the scavenged particles behind for later electron microscopy. The number of drops collected is determined from the residue areas left by the drops. The drop size is determined from photographs from the camera associated with the diffusion chamber.

Another experiment using the same chamber is to work with single large (0.5-8mm) droplets that are positioned in the central volume of the chamber with a syringe. To get the drop into position may require a special suspension system in space. The simple electrostatic device developed by Straubel at Batelle Inst., Frankfurt for suspending drops and particles for various types of measurements would be appropriate. The slight, normal charge on a drop would be sufficient in space to stabilize the drop at a particular location using the oscillating field. Then the field can be turned off and if need be the drop can be repositioned in case of drifting. In the earth-bound laboratory a larger charge has to be used and the field left on to

carry out the same experiment. Once the drop or drops are suspended the relative humidity conditions can be adjusted as desired. For operations below 0 C some care must be taken that the suspension ring will not frost by coating it with glycerine. The ring will be far enough from the drop so it will not affect the ambient conditions near the drop. One can then proceed and expose the drop to the desired aerosol for a desired time increment. Then the drop has to be examined for the amount of aerosol collected. One can use the suspension system to eject the drop onto a collecting surface, such as an electron microscope grid, microscope slide or pulled onto a grid as described above. Finally, one can use the suspension system to determine the mass of the residue by the force needed to hold it suspended in a neutral position with the electrostatic system. In other words the drop is allowed to evaporate while suspended and then the residue is weighed in situ. However, the residue has to be large enough so it can be seen optically.

Both experiments involved techniques that have been used but not in the proposed combination. So it is somewhat presumptuous to evaluate the effort required to bring them to zero-gravity laboratory compatibility. First, the applicability of the approaches should be verified under earth-bound conditions.

Appendix G  
CLASS 7  
RIMING AND AGGREGATION

G.1 RIMING AND AGGREGATION APPROACH 1

G.1.1 Introduction

Results from field experiments points to the existence of riming to be associated with the presence of secondary ice particles in clouds. This fact has led Mossop et al., (1970, 1972) to suggest that riming is a necessary condition for ice multiplication. If water droplets striking and shattering an ice crystal, and throws off ice splinters during the riming process, then it is likely that the charge generation mechanism is also closely associated with riming (Mason, 1971). Apart from its importance as a mechanism for ice multiplication and electrification, riming and aggregation form the basis of precipitation processes. Once an ice crystal growing by diffusion becomes appreciably larger in a dense cloud than the cloud droplets, and has a velocity greater than the ice crystal then the growth of the ice crystal is greatly accelerated by the accretion of these water droplets. This aggregation leads to the formation of graupel which melt later to form raindrops. Theoretical calculations by Ludlam (1952) has shown the need for this type of aggregation for precipitation from clouds warmer than  $-10^{\circ}\text{C}$ . The growth and structure of hailstones by accretion are well understood due to the work mainly by Ludlam (1950), List (1960, 1963), Browning (1966), and Bailey and Macklin (1968). However the physical processes that take place from the time of contact of water droplets and the ice surface to its eventual freezing are not understood. This understanding may give us useful information on the ice multiplication and electrification processes in clouds.

### G. 1. 2 Objective

To determine the mechanism and the processes that take place during the interaction between a supercooled droplet and an ice surface, during events associated with riming and graupel formation.

### G. 1. 3 Scientific Justification

The philosophy of weather modification is based on the fact that there are insufficient ice crystals in a cloud for the release of precipitation. Mason (1955) showed that the number of ice crystals must be  $10^2$  to  $10^3$  times higher than that of the ice nuclei concentration at  $-15^\circ\text{C}$ . This deficiency may therefore be rectified by adding artificial ice nuclei. On the other hand ice crystal concentration measured in certain cumulus clouds indicate that certain natural processes are existing in these clouds to generate ice crystals far in excess than those expected. This raises the fundamental question as to the necessity of knowing which clouds and the condition that are conducive to the production of secondary ice crystals so as to make a decision as to seed or not. Without this knowledge it will not be possible to know whether a cloud requires seeding to make it rain and how much seeding material is required. Furthermore the danger of overseeding a cloud thereby reducing the chances of precipitation, may be avoided if rain augmentation is the objective or encouraged if reduction of rainfall is required.

A poorly understood aspect of cloud physics is the generation of electricity. Since most severe weather phenomena is associated with electricity more evidence is coming to light that electric field itself could affect the precipitation and cloud dynamics. Therefore information as to the importance of various cloud processes to the generation of electrification is a prerequisite to the study of the importance of cloud electrification on the cloud life cycle.

### G. 1. 4 Applications

- 1) Weather modification
- 2) Lightning, forest fires and thunderstorm electrification
- 3) Severe storm processes
- 4) Physics of droplet freezing

#### G.1.5 Terrestrial Laboratory Limitations

All riming experiments are performed with droplets impinging on a stationary or rotating ice surface within a subcooled water cloud. These experiments are useful to study the macroscopic effect of riming on say the total increase in the ice splinters or the charge transferred to the ice surface. Hence the results obtained is rather an average phenomena. On the other hand there is no way to make a microscopic study of the ice-water interaction or study the processes that take place on the drop prior to freezing such as the postulation that the supercooled drop moves on the ice surface prior to freezing.

#### G.1.6 Zero-Gravity Opportunities

The limitations of laboratory experiments to bring a water droplet slowly towards an ice surface are non existent in a low gravity environment. Here a droplet can be focussed to a particular area of the ice surface and observed under a microscope. The influence of gravity on the processes that take place during freezing are not affected by gravity.

#### G.1.7 Quantification

The results anticipated from this experiment may be uniquely achievable only in a low gravity environment.

#### G.1.8 Approach

##### G.1.8.1 General

The experiment consists of three different parts

- 1) The detailed observation of the freezing process.
- 2) The search for any ice splinters that may have been ejected during the freezing and
- 3) The charge transfer (if any) between ice splinters that may have been ejected from the ice surface.

This part needs a microscope for observation. Therefore the cloud chamber should have a small dimension. The crystal has to be located within the working distance of the microscope. Therefore the following preliminary experiment may be considered.

A drop is injected with very low velocity onto an ice surface. The droplet-surface interaction is examined by successive strobe photography using a microscope. The microscope views the drop interactions edge on. The droplet trajectory will be studied by stroboscope and the interaction by direct photography.

These studies, carried out at different temperatures, will give valuable information on the nature of the "liquid" layer on the ice. Precautions must be taken to use high-purity, single-crystal ice. Initial laboratory studies should examine processes occurring as a suspended supercooled drop is brought close to an ice surface at constant velocity; highspeed camera studies may also be required.

In order to incorporate this experiment into the zero-gravity CPL the position of the ice crystal in the SDI must be within about 10 cm of the chamber wall. The detailed interaction of the drop with the ice surface is of interest; namely velocity of approach, instant of freezing and the freezing mechanism itself.

The second part of the experiment requires that the ejected crystals be allowed to grow to a size so as to be observed. Growth of crystals will always occur because the environment will be above ice saturation.

By establishing an electric field before and after the ice-droplet interaction and noting the movement of the ice crystal the charge transferred may be inferred.

The approach therefore will be to use a SDI, place an ice crystal in it taking care to minimize or eliminate any initial charge within the working distance of a microscope. An electric field will then be applied to measure the charge or to verify that it is uncharged. An uncharged droplet is then injected at a relatively low speed and the freezing process observed and photographed using a movie camera attached to the microscope. Any ice crystals that are ejected will be visually counted and photographed. Finally an electric field is applied across the parent crystal to determine the charge.



### G. 1. 8. 2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-crystal	4
Size-droplet	4
Type	
Pollutant	
Pressure	3
Temperature	4
Relative humidity	4
Charge	
Rate of cooling	
Time	
Sound	
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	4
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### G.1.9 Procedure

General test details are given below followed by a representative time line. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure G-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● Unassigned</li> <li>● Personal hygiene</li> <li>● Food</li> <li>● Maintenance</li> <li>● R &amp; R</li> <li>● Sleep</li> <li>● Mission briefing</li> <li>● Subsystem checkout</li> <li>● Preparation               <ul style="list-style-type: none"> <li>Set up experiment</li> <li>Control program</li> <li>Check out experiment</li> <li>Operations</li> <li>Purge differential chamber</li> <li>Calibration</li> <li>Observation</li> <li>Data evaluation</li> <li>Clean up and shut down</li> </ul> </li> </ul>	12 18 30 48 48 480 30 30  42 18 18  1/2 p/ev. 5 3 p/ev. 45 10
<ul style="list-style-type: none"> <li>● Operations breakdown               <ul style="list-style-type: none"> <li>Purge chamber</li> <li>Establish temp</li> <li>Establish RH</li> <li>Establish pressure</li> <li>Inject and position ice crystal</li> <li>Measure temperature of ice crystal</li> <li>Establish electric field</li> <li>Measure charge of ice crystal</li> <li>Remove electric field</li> <li>Project droplet</li> <li>Measure velocity of droplet</li> <li>Measure temperature of droplet</li> <li>Observe interaction</li> <li>Count ice splinters</li> <li>Establish electric field</li> <li>Measure charge of ice crystal</li> <li>Data dump</li> </ul> </li> </ul>	 5 10 10 10 3 2 2 3 1 2 2 5 5 2 2 3 0.5

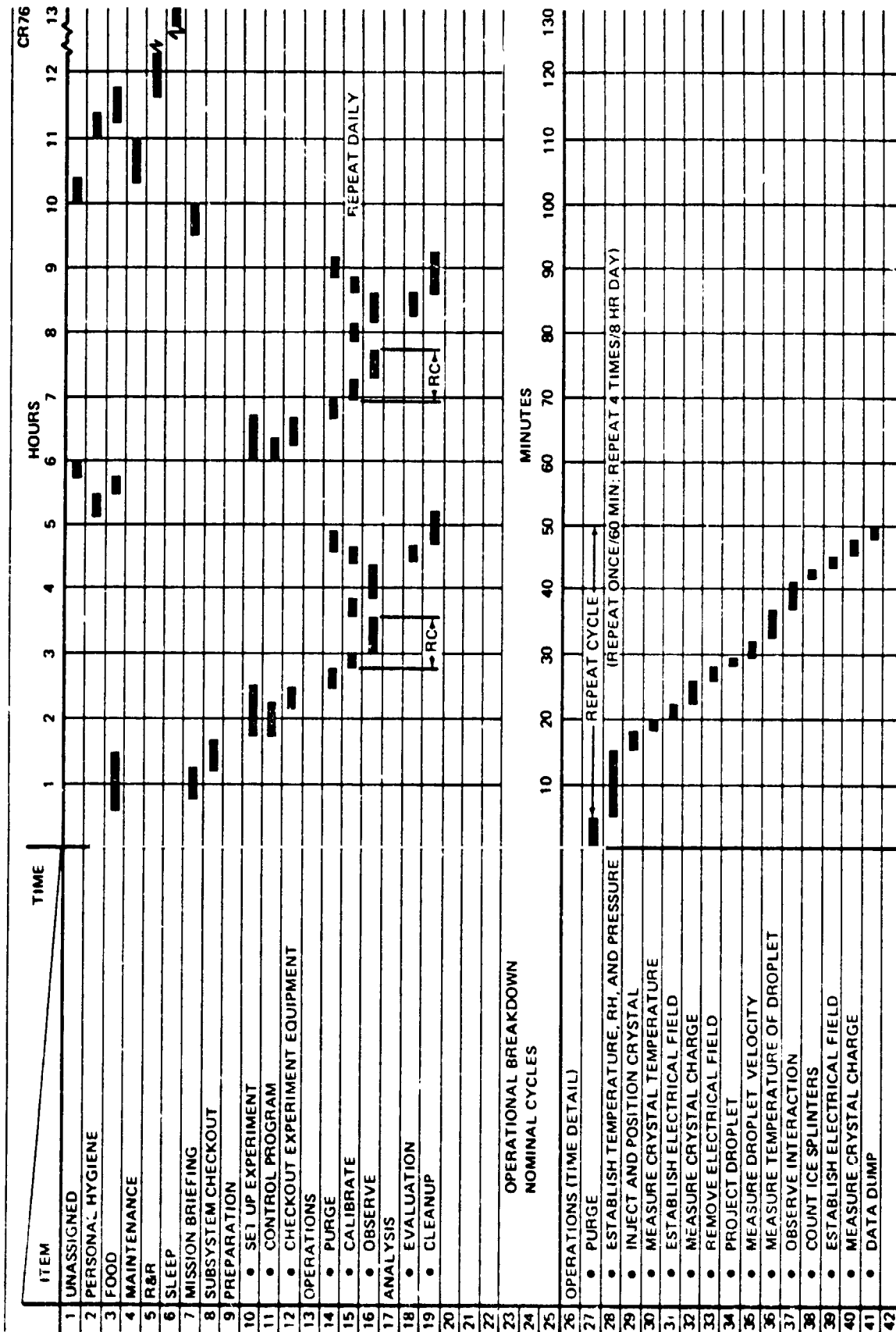


Figure G-1. Activity Timeline (One Day) Experiment Class 7

#### G. 1.10 References

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## G.2 RIMING AND AGGREGATION APPROACH 2

### G.2.1 Introduction

An ice particle falling through a cloud will grow by collecting supercooled drops and/or ice crystals. This collection by an ice particle is a two step process:

- a) aerodynamic capture that brings the drop or ice crystal in contact with the particle surface
- b) attachment to the surface

For ice crystals the overall collection efficiency has been determined by Hosler and Halgren (1) and Latham and Mason (2). These last authors have also found that electric field influences the results.

The adhesion of ice to ice have been studied by Nakaya and Matsumoto (3), Hosler et al (4) and Hobbs and Mason (5).

For supercooled drops it is assumed that they are captured by aerodynamic forces the same as for water drops. The only problem is the choice of the idealized shape to represent the collecting ice particles. The adhesion is supposed to be 100 percent.

It would be desirable to test the accuracy of this assumption and at the same time to study other related problems later, the trajectory of the droplet after impingement on the ice surface, and the effects observed by Hobbs and Farber (6), i. e.,

- a) breaking a branch of an ice particle because of the energy of the drop impaction
- b) breaking a branch of an ice particle because of localized heating due to the released freezing heat.

### G.2.2 Objective

The major objective of the experiment is to study the interaction of supercooled drops and an ice surface. The main purpose is to establish how the adhesion to the surface depends on the external conditions.

### G.2.3 Scientific Justification

Theories of precipitation involving supercooled clouds rely on the collection of supercooled drops by ice particles. A better knowledge of this process will make the models more realistic.

Another aspect to be considered in connection with the theory of precipitation is the ice particles multiplication. Besides the splinters and fragments produced when the drop freezes, the two mechanisms mentioned Hobbs and Farber (6) can be relevant in appropriate conditions.

A third type of problem is the electrification of clouds. How a falling ice particle becomes electrically charged when it collects supercooled drops has been studied by Reynolds (7), Reynolds, Brook and Gourley (8), Magono and Takahashi (9), Latham and Mason (2).

A better knowledge of the interaction of supercooled drops and ice particles will improve our knowledge in this field.

### G.2.4 Applications

The main fields in which this knowledge can be applied are: Cloud Physics, Weather Modification, Cloud Electrification and Icing on Flying and Stationary Objects.

### G.2.5 Terrestrial Laboratory Limitations

The main limitations of the terrestrial experiments are the need of a support for the ice particles and that the velocities at which the supercooled drops were projected not always matched the velocities in the real situation.

### G.2.6 Zero-Gravity Opportunities

The absence of gravity eliminates the presence of support for the ice crystal, specially for the small ice particles.

In zero gravity conditions the interaction of individual drops and the ice surface can be followed. The movements of the drops before attachment or not getting attached, their freezing patterns and their interaction in ice multiplication mechanisms.

As an overall result, the efficiency of the adhesion to the ice surface can be determined, filling an important gap in the theory of precipitation.

#### G.2.7 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure G-2).



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● Stabilize the SDI for the particular temperature range</li> <li>● Inject ice crystal germ</li> <li>● Grow the ice crystal</li> <li>● Inject drop</li> <li>● When drop is injected, start taking pictures and continue to do so during the next 5 minutes taking a picture every 10 seconds</li> <li>● Remove all the particles in the chamber</li> <li>● Repeat</li> </ul>	<div>15</div> <div>1</div> <div>3</div> <div>1</div> <div>5</div> <div>3</div>

### Experiment Parameters

The important experiment parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-crystal	5
Size-droplet	3
Type	5
Pollutant	
Pressure	3
Temperature	4
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

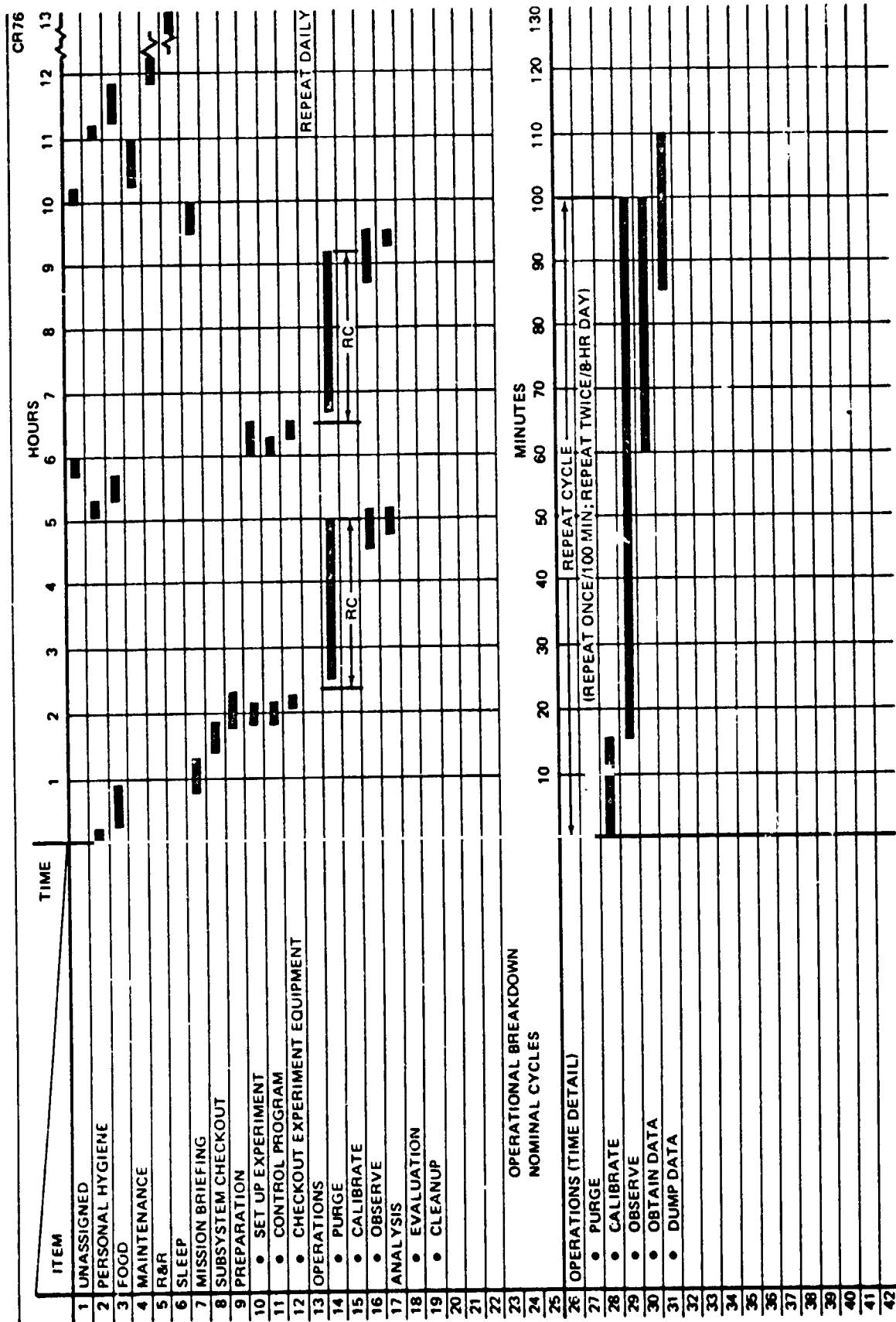


Figure G-2. Activity Timeline (One Day) Experiment Class 7

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Appendix H  
CLASS 8  
DROPLET-ICE CLOUD INTERACTIONS

## H.1 INTRODUCTION

In addition to the suggested approach in the September 1973 report the following may prove to be useful. The object is to introduce an accurately known number of ice nuclei into a supercooled cloud and then to count all the resulting ice crystals. The best approach here is to introduce the nuclei in the form of very small ice crystals — in the 1 micron range. These crystals could be generated by overseeding and then transferring a small sample into the test chamber. A similar sample should be counted for the ice crystal concentration. For the latter purpose a modified NCAR ice nucleus counter could be used, i. e., in space the size of the instrument could be reduced because one is not concerned with settling losses, which require continuous flow on the ground to obtain useful data. As for the test chamber, it is envisioned that the device would be operated in a batchwise manner or more probable at a very low velocity and then the whole sample would be aspirated through an acoustic counter. This procedure would show how many additional crystals are formed by multiplication processes under a variety of conditions. It would not give information on how the process took place, so the experiment should be supplemented by photographic work. This will be more difficult and the above would serve as a screening process to pick out the most interesting cases. Then the focus can be on individual events not the overall results.

The following is from the NASA CR-12903 September 1973 report.

## H.2 OBJECTIVE

In the proposed experiment, determine the modes and extent of the interactions of ice crystals and supercooled water droplets, including the propagation of the ice phase through a supercooled droplet cloud and the diffusional

growth of ice crystals within a cloud of supercooled droplets under varying conditions of temperature, pressure, and droplet/crystal concentrations.

### H.3 SCIENTIFIC JUSTIFICATION

The propagation of ice throughout the upper levels of clouds (e. g., thunderstorm cumuli) has extreme relevance to all attempts to modify weather. Aircraft observations have indicated that the numbers of ice nuclei near a cloud base are often several decades lower than the number of nuclei necessary to explain the rapidity with which the ice phase moves through the upper parts of a supercooled cloud as determined by radar. An explanation of this rapid propagation of the ice phase is the multiplication of ice particles (e. g., by droplet breakup upon freezing and ice crystal breakup during collisions). These fragments then serve as ice nuclei resulting in the cascading of the ice phase through the cloud. The extent and conditions for natural ice nuclei production must be known before decisions can be made concerning the quantity of seeding material that is injecting for a specific modification objective. For some rain and snow processes, too many nuclei cause competition for the available water among the generated ice crystals. This results in small crystals which have less probability of forming rain-size precipitation. At the other extreme, too few seeding nuclei would not release the thermodynamic energy of a precipitation system that would result in precipitation. Each precipitation process has different seeding requirements, and a knowledge of the total natural and man-injected nuclei properties must be available.

Another important aspect of the cold precipitation process is the growth of ice crystals within a cloud of supercooled water droplets. The crystal types and rate of growth of multiparticles must be studied. Such studies will provide an indication of the conditions and times under which seeding must be done to have maximum effectiveness for a specific weather modification goal.

The two examples above are representative of the many processes which take place during the ice phase of precipitation growth. Each aspect of these processes must be studied separately involving only a few particles under very controlled laboratory conditions before a process can be fully understood

in relation to its complex interaction with the environment. For this reason, many of the classes of experiments proposed for zero-gravity, as in a terrestrial laboratory, deal with single or few particles to isolate specific processes for detailed studies.

A necessary step in understanding the complete atmospheric precipitation process is to simulate a large parcel of particles for the study and observation of several microphysical processes proceeding simultaneously. It is this latter aspect to which this experiment is directed. Once individual processes such as diffusion growth and conditions for droplet splintering are understood, then the complex interaction studies can better be approached. Effects of sound, optical, and electrical fields will also be studied in relation to the ice-droplet interactions.

These experiments also will be used to provide knowledge as to the extent to which inadvertent weather modification takes place due to man (e. g. , through pollution and urban development).

#### H.4 APPLICATIONS

The growth of ice crystals and their propagation is important in all attempts to modify cold precipitation processes (e. g. , snow, hail, sleet, and thunderstorms). The generation of natural ice crystal nuclei has a bearing on the quantity of nuclei that may be needed to achieve a specific weather modification goal. Overseeding and underseeding can defeat the original objective.

#### H.5 ZERO-GRAVITY OPPORTUNITIES

Observations in the terrestrial laboratory are limited by particle fallout and to some extent convection, which are both gravity-driven. Observation times are limited to milliseconds in small expansion chambers, seconds in diffusion chambers, and tens of seconds in very large chambers. With the large chambers, convection prevents the continuous observation of specific particles, while for the smaller chambers, the seconds available are not enough compared to the minutes available within natural atmospheric clouds. A low-gravity environment would permit the observation of individual crystals and droplets for times that are representative for atmosphere relevant processes.

## H.6 APPROACH

### H.6.1 General

The nucleation of ice and ice crystal growth within a supercooled cloud of droplets are used as representative experiments of this class.

A cloud of supercooled droplets will be injected into a thermally controlled chamber. Ice or water surfaces will provide an appropriate humidity controlled environment. For a number of the observations the chamber will be raised to near saturation at or above freezing temperature by the use of a purge and humidification system. A cloud of water droplets will then be injected into the chamber. As the chamber is cooled below freezing, the air will become saturated and the supercooled droplets will grow or evaporate as a function of the initial relative humidity and pressure, as well as the final temperature and pressure. At a selected final condition of temperature and relative humidity, some ice nuclei will be injected to produce a few ice crystals. As the supercooled droplets freeze, visual and photographic observations will be made of the freezing of adjacent droplets and the resulting ice phase propagation, as a consequence of droplet splintering (ice multiplication). The application of sound, optical, and electric fields will also be studied in relation to their influence on the ice phase propagation (e.g., due to electric field-driven charged particles generated by freezing supercooled droplets). Freezing could be initiated by other means including a cold probe or dry ice in addition to the above-mentioned ice nuclei.

Observations will also be made of the ice crystal type and growth rate at the expense of adjacent supercooled droplets as a function of ambient pressure, temperature, and relative humidity. These observations will include, e.g., the rates of evaporation of adjacent droplet and a measure of the sphere of influence for an ice crystal (i.e., that volume from which an ice crystal draws water vapor at the expense of the surrounding supercooled water droplets). The variables of interest include droplet and crystal concentrations and sizes, rates of growth, ambient temperature, pressure, relative humidity and magnitude of electric, sound and optical fields.



### H.6.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

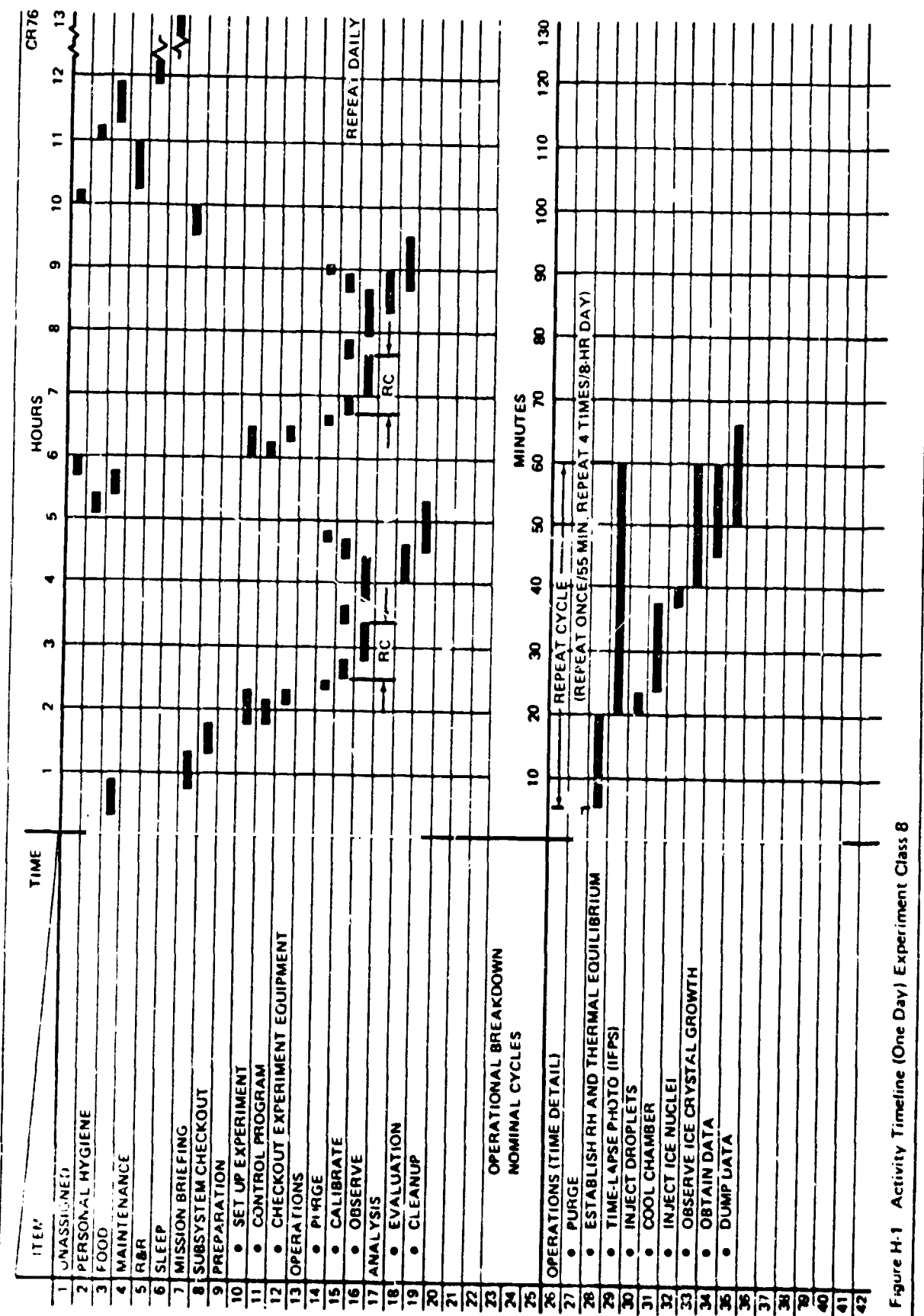
<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	4
Type	6
Pollutant	
Pressure	3
Temperature	4
Relative humidity	
Charge	4
Rate of cooling	
Time	6
Sound	3
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	3
Ventilation	
Optical	4
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

## H.7 PROCEDURE

General schedule details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations to translating this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure H-1).

PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Purge chamber</li> <li>• Establish humidity and thermal equilibrium (+5°C)</li> <li>• Start time-lapse photographs (1/second)</li> <li>• Inject droplets (1 to 1,000 per cm<sup>3</sup>)</li> <li>• Cool chamber to subfreezing temperature</li> <li>• Inject ice nuclei</li> <li>• Visually observe freezing and growth of ice crystals</li> <li>• Stop camera</li> <li>• Recycle to other final relative humidity and temperature</li> <li>• Recycle other droplet sizes</li> <li>• Recycle other droplet/crystal concentrations</li> <li>• Recycle with electric, optical, sound fields</li> </ul>	<p>5</p> <p>15</p> <p>3</p> <p>15</p> <p>3</p> <p>20</p>



Appendix I  
CLASS 9  
HOMOGENEOUS NUCLEATION (ICE)

I. 1 HOMOGENEOUS NUCLEATION (ICE) APPROACH 1

I. 1. 1 Introduction

Although homogeneous ice nucleation normally does not take place in the atmosphere, the concepts established through its study can be applied to other nucleation processes. In this regard, quantitative and reliable data of the homogeneous ice nucleation, as it is the simplest of all the ice nucleation modes, will help understand more complex ice nucleation processes.

Homogeneous ice nucleation is a function of time and temperature, as well as of the characteristics of the pure water such as the surface free energy of the ice-water interface. A possible direct application for results of homogeneous ice nucleation experiments is the determination of this interfacial free energy which cannot be determined in any other known way. Various physical support, such as solid or liquid surfaces, and errors in droplet temperature determinations due to free fall have made it impossible to accurately estimate this quantity.

However, the low gravity condition of the space laboratory provides a unique opportunity of avoiding such difficulties and permits one to obtain accurate data concerning homogeneous freezing of water droplets.

I. 1. 2 Objective

The main objectives of this study are first to determine the homogeneous freezing rate of pure supercooled water droplets as a function of time, temperature and droplet size, and second to estimate the surface free energy in ice-water interface. The first part is experimental.

I. 1. 3 Scientific Justification

The justification for conducting this experiment is stated in the introduction.

#### I. 1. 4 Applications

The experiment applications are stated in the introduction.

#### I. 1. 5 Terrestrial Laboratory Limitations

The terrestrial laboratory limitation of this research subject originates from the difficulty of avoiding influences from supporting surfaces. Use of the falling droplet method allows one to obtain a supporting surface free condition, however, a new problem arises instead which is based on the uncertainty of droplet temperature during dropping.

#### I. 1. 6 Zero-Gravity Opportunities

The low gravity condition of the space laboratory provides a unique possibility of suspending large droplets without any support, and enables us to evaluate more accurately the homogeneous freezing behavior of supercooled droplets.

#### I. 1. 7 Approach

##### I. 1. 7. 1 General

The principle of this study is to suspend droplets in the median plane of the SDI chamber at a temperature slightly above that of investigation and under water saturation. Then, the temperature will be lowered very slowly. The number of frozen droplets, the temperatures of top ( $T_1$ ) and bottom ( $T_2$ ) plates as well as that of median plane ( $T_M$ ) will be recorded with respect to time.

The experimental procedure is as follows. Droplets will be prepared by two methods. To produce droplets smaller than 100  $\mu\text{m}$  in diameter, the clean chamber air will be adiabatically cooled under  $\Delta T = T_1 - T_2 = 0$ , above  $0^\circ\text{C}$  so that the homogeneous condensation will take place. The formed fog will be diluted with clean nuclei free air to the desired concentration. Then, by giving a suitable  $\Delta T$ , the droplets will be permitted to grow to the final size. The formed droplets are clean and free of nuclei. For production of droplets larger than 100  $\mu\text{m}$  in diameter, a vibrating needle droplet generator will be used. A few sizes of needles will be needed depending on the range of droplet diameters. Water to be used must be very clean. Droplets will

be sent along the median plane of the SDI Chamber. The viscous resistance of chamber air will stop droplets, and they will be suspended in the plane.

When droplets are produced in the chamber, the chamber will be cooled by a relatively rapid expansion to a temperature slightly above that for study. During this process,  $T_1$  and  $T_2$  will be lowered allowing  $T_M$  to follow the temperature of expansion and to hold water saturation in the median plane. From this point on, a very slow cooling will proceed. The median plane of the chamber will be illuminated with a laser beam and photomicrographs will be taken at regular intervals. The time,  $T_1$ ,  $T_2$ , and  $T_M$  will be recorded.

#### I. 1. 7. 2 Instrumentation

Apart from an ordinary supporting facility to produce very clean filtered air, major instruments to be used are as follows. The SDI Chamber will be used with minor modifications to carry out the adiabatic expansion and to introduce droplets. A vibrating needle droplet generator, with interchangeable needles of different sizes and capable of changing the frequency and amplitude, will have to be developed. For recording, a multichannel stripchart recorder of temperature and pressure and a microscope of long working distance with a camera will be needed.

### 1.1.7.3 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	10
Type	
Pollutant	
Pressure	
Temperature	5
Relative humidity	
Charge	
Rate of cooling	5
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	



### **I. 1. 8 Procedure**

General activity details are given below followed by a representative activity timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations of this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient (see Figure I-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Purge SDI chamber</li> <li>• Expand chamber air for homogeneous condensation</li> <li>• Bring pressure back slowly to 1 atm, dilute fog</li> <li>• Grow fog droplets</li> <li>• Generate droplets by vibrating needle apparatus</li> <li>• Cool chamber adiabatically and relatively rapidly to temperature slightly above that for study</li> <li>• Activate laser</li> <li>• Slowly cool chamber</li> <li>• Photograph frozen droplets</li> <li>• Record <math>T_1</math>, <math>T_2</math>, <math>T_M</math></li> <li>• Recycle at other temperatures for study</li> <li>• Recycle at other droplet sizes</li> </ul>	<div>5</div> <div>20</div> <div>5</div> <div>5</div> <div>2</div> <div>15</div>

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● Purge SDI chamber</li> </ul>	5
<ul style="list-style-type: none"> <li>● Expand chamber air for homogeneous condensation Bring pressure back slowly to 1 atm, dilute fog Grow fog droplets</li> </ul>	20
<ul style="list-style-type: none"> <li>● Generate droplets by vibrating needle apparatus</li> </ul>	5
<ul style="list-style-type: none"> <li>● Cool chamber adiabatically and relatively rapidly to temperature slightly above that for study</li> </ul>	5
<ul style="list-style-type: none"> <li>● Activate laser</li> </ul>	2
<ul style="list-style-type: none"> <li>● Slowly cool chamber</li> </ul>	15
<ul style="list-style-type: none"> <li>● Photograph frozen droplets</li> </ul>	
<ul style="list-style-type: none"> <li>● Record <math>T_1</math>, <math>T_2</math>, <math>T_M</math></li> </ul>	
<ul style="list-style-type: none"> <li>● Recycle at other temperatures for study</li> </ul>	
<ul style="list-style-type: none"> <li>● Recycle at other droplet sizes</li> </ul>	

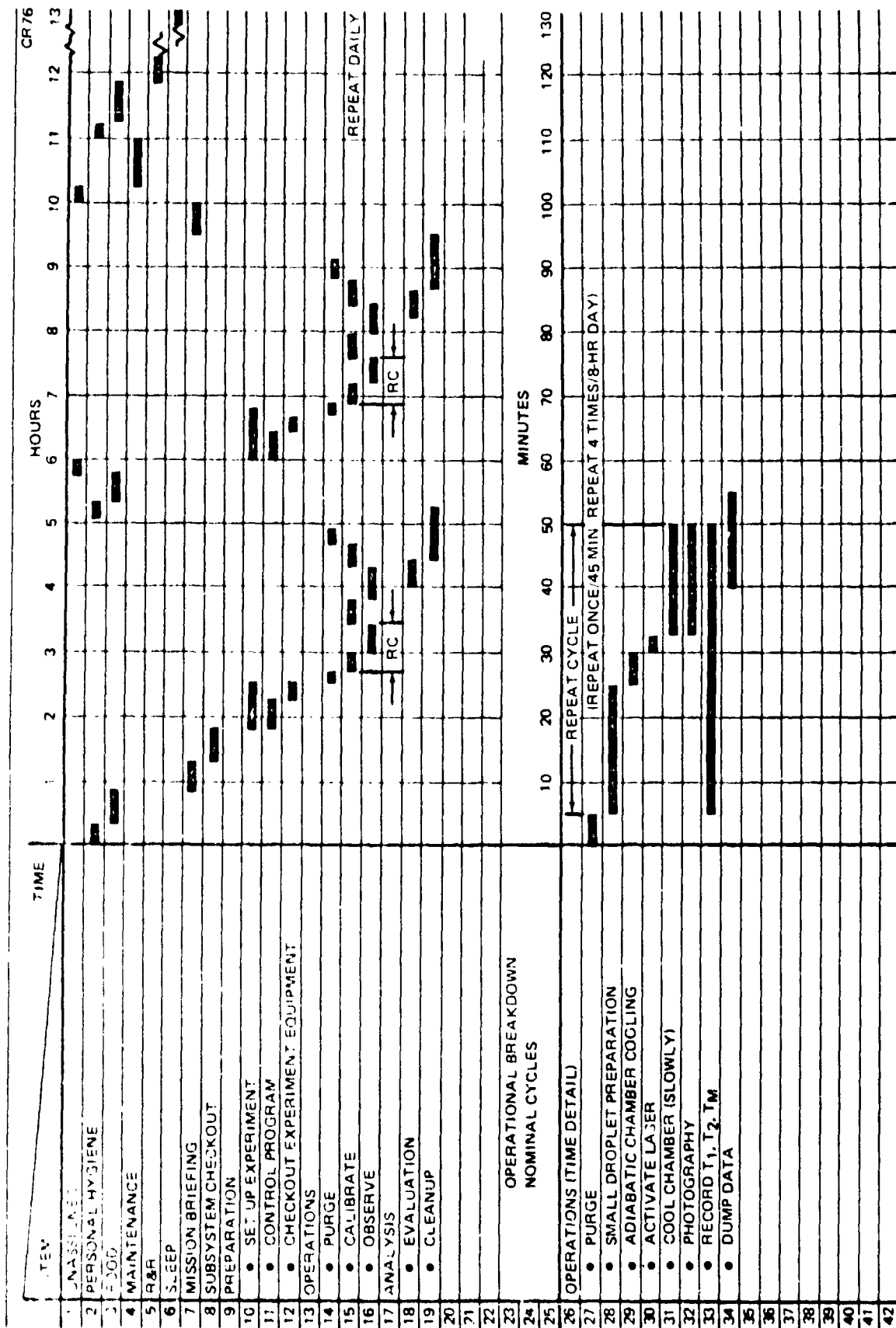


Figure 1-1. Activity Timeline (One Day) Experiment Class 9

## 1.2 HOMOGENEOUS NUCLEATION (ICE) APPROACH 2

### 1.2.1 Introduction

It is well established that water droplets can exist in a supercooled state for temperatures well below  $0^{\circ}\text{C}$ . Most indications are that  $-40^{\circ}\text{C}$  is the lower limit at which a small supercooled droplet exists before it will spontaneously freeze. In the atmosphere the extent to which a drop can supercool is determined by the presence of ice nuclei and the phenomena of homogeneous nucleation is only of theoretical interest while the question of heterogeneous freezing is very germane to weather modification. The exception for this is the formation of ice fog in arctic cities e.g. Fairbanks, Alaska when the temperature falls below  $-35^{\circ}\text{C}$  when evaporation of warm water surfaces leads to spontaneous formation of ice crystals. In these situations homogeneous nucleations are of great interest.

In order to understand heterogeneous nucleation, a clear understanding of the homogeneous nucleation of ice crystals is very necessary. The thermodynamic processes that take place in heterogeneous nucleations follow logically from the theories developed in homogeneous nucleation (see Fletcher 1962). Hence sets the framework for the study of homogeneous nucleation.

Early experiments on homogeneous nucleation by Bigg (1953), Mossop (1955), Langham and Mason (1958) and many others indicate that it is statistical phenomena and the median temperature of spontaneous freezing increases as the drop diameter increase (see Mason 1971 for a critical discussion). The other important factor is the cooling rate. Here the effect is small giving an increase of about  $0.5^{\circ}\text{C}$  in the median temperature for a change from  $0.05\text{ C/min}$  to  $0.5\text{ C/min}$  in the cooling rate.

These early experiments on supercooling were performed with drops supported either on a solid surface or between immiscible liquids. Hence doubts remain for their validity in free air. For this reason Kuhns and Mason (1969) carried out experiments on the freezing of freely falling droplets of very pure, particle free water. But since no control of the cooling rate was possible their results have to be corrected first for the thermal lag and then standardized to a particular cooling rate for comparison with

other results. Most of these results compare favorably with the theory of homogeneous nucleation but all laboratory experiments need some kind of correction to account for the uncertainties of the influence of the suspension device or the temperature of the drop.

### I. 2. 2 Objective

Determine the homogeneous freezing distribution of very pure, nuclei free water droplets as a function of degree of supercooling, droplet diameter, the cooling rate and the composition of the environment under conditions of no physical support.

### I. 2. 3 Scientific Justification

The formation and behavior of clouds are, in part, regulated by the micro-physical processes active in the formation, development, and behavior of the individual droplets. The mechanisms by which atmospheric nuclei become activated and grow to cloud size droplets is not completely understood. The heterogeneous nucleation process is complicated by the presence of a foreign particle which is usually of unknown composition and possesses surface properties which are not readily characterized. Even the simplest of nucleation processes, the homogeneous nucleation of liquid droplets from the vapor, is not fully understood. Although homogeneous nucleation does not occur in the atmosphere, the concepts established through its study will form a foundation upon which an understanding of the heterogeneous nucleation process can be developed. Therefore, a quantitative understanding of the homogeneous nucleation process must precede our comprehension of the more complex heterogeneous nucleation process.

Homogeneous nucleation is a function of time and temperature, as well as of the characteristics of the "pure" water such as the specific surface free energy of the crystal/liquid interface. A possible result of homogeneous nucleation experiments is the determination of the free energy of this crystal/liquid interface which cannot be accurately measured or calculated in any other way. Various physical supports and errors in droplet surface temperature determinations due to free fall have not yet given a reliable determination of this quantity.

#### I. 2. 4 Applications

- A. Low temperature fog e. g. ice fog
- B. Contrail formation
- C. The physics of nucleation

#### I. 2. 5 Terrestrial Laboratory Limitations

In the experiments designed to study the objectives listed above, terrestrial laboratory experiments are limited by the suspension device or the uncertainty of the temperature. Many of the homogeneous nucleation experiments have been performed with the water droplets on polished metallic plates between two immiscible liquids or with the liquid sealed within the glass or quartz tube. All of these methods involve surface contact which can modify the surface free energy of the droplet and, thus, the support media can serve as nucleation sites. Of these methods, the two liquid approach appears to be the best. In the other types of terrestrial experiments where drops are allowed to fall freely, these experiments are limited to small droplets falling through small temperature gradients so as to achieve the terminal velocity and temperature equilibrium of the drop. However there is uncertainty in the assumption of the temperature of the drop has reached the environment temperature. Although corrections may be used for these effects, erroneous conclusions may be reached regarding the mechanism of homogeneous nucleation and the values of the important parameters such as surface energy when attempts are made to relate the experiment results to the theory.

#### I. 2. 6 Zero-Gravity Opportunities

A potential solution to the number of experimental problems encountered in terrestrial experiments in the free suspension of droplets in air under low gravity conditions. These experiments can then be performed over extended time periods with slow rates of cooling to eliminate thermal time lag problems and performed without surface contact with a foreign material.

#### I. 2. 7 Approach

##### I. 2. 7. 1 General

A cloud of pure water droplets will be injected into a thermally controlled chamber while the chamber is above freezing (e. g. , +5°C). Then the chamber

will be cooled slowly, less than  $0.5^{\circ}\text{C}$  per minute, and photographic data taken at  $0.1^{\circ}\text{C}$  intervals. Because of volume of data required, holographic techniques would be ideally suited. These data would provide sizes and numbers of droplets and crystals versus time, temperature, and rate of cooling. Cooling continues until all droplets have frozen which will take place by about  $-40^{\circ}\text{C}$  depending on droplet characteristics (such as volume). Surface area and volume dependence of this statistical freezing will be studied by inserting clouds which all have the same total liquid volume but different droplet diameter (i. e., different surface areas). Consideration must be given to the maximum allowable droplet/crystal density that will avoid diffusion interaction of adjacent particles. Other clouds with the same surface areas but different total liquid volumes will provide further information concerning volume and surface area dependence of homogeneous freezing. At present, most results favor the volume dependence, but experimental uncertainties and difficulties leave room for some question about this.

#### I. 2. 7. 2 Instrumentation

The thermally controlled chamber will be capable of being slowly and uniformly cooled to temperatures as low as  $-40^{\circ}\text{C}$ . Special precautions must be taken to prevent ice nucleation on the walls of the chamber. Non-nucleating fluids show the most promise below  $-20^{\circ}\text{C}$ , while special teflon surfaces are adequate above this temperature. Droplet injection techniques are available which permit placement of individual droplets within a few millimeters of a desired location. In this way, a single layer of droplets, appropriately spaced to prevent interactions, can be provided to conform to the depth of field limitations of a normal camera system. Holographic techniques would remove this restriction, permitting much more information to be collected at a given time because of its volume recording capabilities.

#### I. 2. 7. 3 Measurement and Data Requirements

Photographic records will provide the basic information on droplet and crystal numbers, spacing, and sizes which is to be correlated with cooling rates, temperature, time, and ambient relative humidity. Vocal recorded commentary will be utilized throughout the experiment along with digital recording of time, temperature, pressure, and relative humidity. Analog displays of these variables will also be available during the experiment.



#### I. 2. 7. 4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	4
Type	
Pollutant	3
Pressure	4
Temperature	
Relative humidity	
Charge	
Rate of cooling	4
Time	6
Sound	3
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	4
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### I. 2. 8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure I-2).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Unassigned	12
• Personal hygiene	18
• Food	30
• Maintenance	48
• R & R	48
• Sleep	480
• Mission briefing	30
• Subsystem checkout	30
• Preparation	
Set up experiment	42
Control program	18
Check out experiment	18
Operations	
Purge differential chamber	1/2 per event
Calibration	5
Observation	3 per event
Data evaluation	45
Clean up and shut down	10
• <u>Operations breakdown</u>	
• Purge	5
• Flush	2
• Purge (very clean)	5
• Introduce ambient gas	5
• Establish pressure	10
• Establish RH (water saturation)	10
• Establish temperature	10

**PROCEDURE**

<b>DETAILED ACTIVITY</b>	<b>TIME REQUIRED MINUTES</b>
<ul style="list-style-type: none"><li>● Inject cloud droplets</li><li>● Proceed cooling</li><li>● Time lapse photography</li><li>● Obtain data</li><li>● Data dump</li></ul>	<p>3</p> <p>40</p> <p>40</p> <p>40</p> <p>1</p>

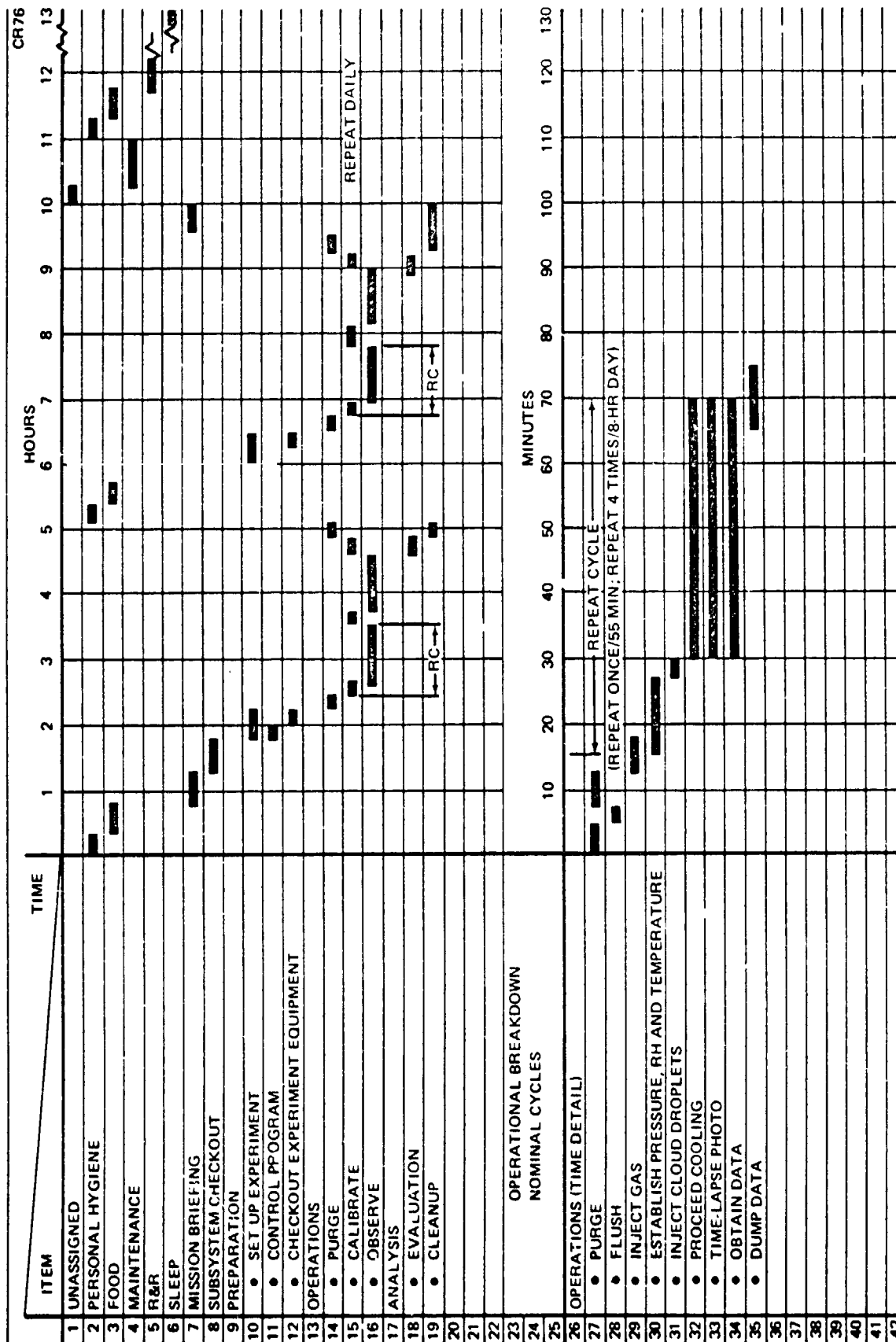


Figure 1-2. Activity Timeline (One Day) Experiment Class 9

### 1.2.9 References

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### I. 3 HOMOGENEOUS NUCLEATION (ICE) APPROACH 3

#### I. 3. 1 Introduction

At times the sparsity of nuclei in the atmosphere which possess the potential for catalyzing the nucleation of ice in cloud results in the supercooling of cloud droplets frequently to temperatures as low as  $-20$  to  $-35^{\circ}\text{C}$  before freezing occurs. In strong thunderstorm updrafts, a scarcity of ice nuclei is believed to be a necessary requirement for the production of hail. Observations of the evolution of natural cloud systems has generated considerable interest in the various mechanisms by which ice may form in cloud. While large masses of water almost always freeze with very little supercooling, water subdivided into small droplets inevitably exhibits a large number of droplets which sustain very substantial supercooling before freezing. Since the degree of supercooling is very sensitive to the concentration of particulate impurities, it is evident that certain particulates possess properties which effectively catalyze the freezing nucleation process. The composition and surface properties of nuclei possessing this special characteristic are unknown. Moreover, even the simplest of freezing processes, homogeneous nucleation, is poorly understood. Although homogeneous freezing nucleation is not likely to occur very often in the atmosphere, the concepts established through its study will help to formulate the theoretical foundations for understanding the more complex heterogeneous nucleation process. Therefore, a quantitative understanding of the homogeneous freezing nucleation process must precede a comprehensive of heterogeneous nucleation.

The formation of the first recognizable fragments of the solid phase in the midst of the liquid phase is energetically unfavorable due to the large surface to volume ratio of the ice embryo and the positive free energy associated with the creation of the new interface. The probability that statistical fluctuations in the local energy will be sufficient to surmount these free energy barrier increases with the degree of supercooling. A systematic study of homogeneous freezing nucleation can result in the determination of the free energy of the solid-liquid interface which cannot be accurately measured or calculated in any other way.

The major problem encountered by experimentalists, in attempting to investigate the homogeneous freezing nucleation of water droplets, has been in devising various means of supporting the droplets for long periods of time while they are being supercooled under the watchful eye of the investigator. Various schemes for supporting the droplets introduce new surfaces which may be a source of freezing nuclei and other impurities. Experiments using freely falling droplets introduce other extraneous effects such as temperature lag and the effect of internal convection on the freezing process which lead to ambiguities in the data. The advent of the zero-g laboratory introduces a new technique which offers the potential for eliminating most of the objections found in terrestrial experiments and offers the potential for obtaining data which is sufficiently consistent to be of real value in developing and evaluating the theory. These experiments augment other experiments proposed for this program in which heterogeneous nucleation will be studied.

#### 1.3.2 Objective

The objective of this experiment is to make systematic observations of homogeneous freezing nucleation in free-floating droplets as a function of time, degree of supercooling and drop size. This data will be used in the development and evaluation of theoretical concepts which are fundamental to understanding the more complex process of heterogeneous freezing nucleation which is of fundamental importance in cloud physics.

#### 1.3.3 Scientific Justification

Large bodies of water almost always are observed to freeze at temperatures only slightly below 0°C due to the abundance of freezing nuclei in ordinary water sources. If however, the water is subdivided into small droplets, the likelihood that each droplet will contain an active freezing nucleus steadily diminishes with decreasing drop size. An appreciable portion of these droplets are observed to supercool substantially below the equilibrium freezing temperature, the droplets remaining in a metastable state. The smaller the concentration of particulate impurities, the greater is the degree of supercooling which can be achieved up to what appears to be a limit. Most liquids appear to exhibit an intrinsic property whereby homogeneous freezing limits the degree of supercooling to about  $0.8 T_F$ , where  $T_F$  is the equilibrium



absolute freezing temperature, regardless of the degree of subdivision of the fluid (Ubbelohde, 1965).

The formation of the first fragments of the solid phase in the presence of the liquid phase is energetically unfavorable because of the large surface to volume ratio and the positive free energy associated with the creation of the new interface (Fletcher, 1970). The free energy required to form a spherical ice embryo of radius  $r$  is

$$\Delta G(r) = \frac{4}{3} \pi r^3 \Delta G_v + 4 \pi r^2 \sigma_{SL} \quad (1)$$

where  $\sigma_{SL}$  is the interfacial free energy and  $\Delta G_v$  is defined as the difference in free energy between the ice and water on a per unit volume basis.

$$\Delta G_v = n_s (\mu_s - \mu_L) \quad (2)$$

where  $n_s$  is the number of molecules in the ice embryo and  $\mu_s$  and  $\mu_L$  are the chemical potentials of the solid and liquid states respectively. The size of the critical embryo,  $r^*$ , can be obtained by setting  $\partial \Delta G / \partial r$  equal to zero.  $\Delta G$  is found to possess a maximum at  $r^*$ .

$$r^* = -2\sigma_{SL} / \Delta G_v \quad (3)$$

The corresponding value of the free energy is  $\Delta G_v^*$

$$\Delta G^* = \frac{16\pi\sigma_{SL}^3}{3(\Delta G_v)^2} \quad (4)$$

The latter assumes that  $\sigma$  is isotropic and that the embryo is spherical. Uncertainties in  $\sigma_{SL}$  are such that further refinements in (4) are not warranted.

Since  $S = -(\partial G/\partial T)$  the average entropy of fusion  $\langle \Delta S_v \rangle$  over range of supercooling  $\Delta T$  is given by:

$$\Delta G_v = \int_0^{\Delta T} \Delta S_v d\Delta T = -\langle \Delta S_v \rangle \Delta T \quad (5)$$

Thus,

$$\Delta G^* = \frac{16 \pi \sigma^3}{3 (\langle \Delta S_v \rangle \Delta T)^2} \quad (6)$$

Thermal fluctuations give rise to an embryo distribution described by a Boltzmann equation

$$n(r^*) = n_L \exp(-\Delta G^*/kT)$$

The nucleation rate is the rate at which embryos of size  $r^*$  gain an additional molecule. Although, in the case of nucleation of liquid drops from the vapor the kinetic coefficient was readily determined from kinetic theory, in the case of the liquid-solid interface things are not so simple. Here it is a case of molecules becoming detached from the liquid phase and becoming properly oriented and handed into the solid phase embryo. The nucleation rate was evaluated by Turnbull and Fisher (1949) and found to be given approximately by

$$j \approx n_L B \exp(-\Delta G^*/kT) \quad (7)$$

where the kinetic coefficient  $B$  is given by

$$B = \frac{kT}{h} \exp(-\Delta g_{SL}/kT) \quad (8)$$

and  $h$  is Planck's constant and  $\Delta g_{SL}$  is the activation energy for self diffusion across the liquid-solid interface. McDonald (1953) finds  $\Delta g_{SL} \approx 4 \times 10^{-13}$  erg/molecule. The factor  $n_L kT/h \approx 10^{35} \text{ cm}^{-3} \text{ sec}^{-1}$  and  $-\Delta G^*/kT \approx 10^{30} \text{ cm}^{-3} \text{ sec}^{-1}$ . Attempts to calculate the degree of supercooling at which

J becomes observable yield widely differing values, indicating only that  $\sigma_{SL}$  lies somewhere between 10 and 25 ergs/cm<sup>2</sup>. Fletcher (1970) estimates the best value of  $\sigma_{SL}$  to be about 22 erg/cm<sup>2</sup> and the size of  $r^*$  to be about 190 molecules at -40°C. Thus, it is evident that one important result of a homogeneous freezing nucleation experiment is that it will allow  $\sigma_{SL}$  to be evaluated experimentally and offer additional information about the magnitude of  $\Delta g_{SL}$ .

Nemethy and Scheraga (1962) applied the techniques of statistical thermodynamics to the flickering cluster model of Frank and Wen (1957) in an effort to develop a statistical mechanical theory of liquid water. Kuhns and Mason extrapolated the most probable cluster size associated with Nemethy and Scheraga's model down to temperatures of the order of -40°C and compared this curve with curves for the number of molecules in the critical cluster which he obtains from their experimental data for several different values of  $\sigma_{SL}$  near  $\sigma_{SL} = 20$  erg/cm<sup>2</sup> which they find match their experimental results. They find that the  $\sigma_{SL} = 20$  erg/cm<sup>2</sup> curve crosses the extension of the Nemethy and Scheraga curve at about -40°C. They regard this result as fortuitous but nevertheless supporting evidence for the flickering cluster model.

Eadie (1967) wrote a dissertation on a molecular theory for the homogeneous freezing nucleation of supercooled water. Although the approach developed by Eadie has not been followed up, Bolander, Kassner and Zung (1969) introduced a molecular model for vapor phase nucleation which they applied to the dimer and trimer clusters in the absence of a generalized method of calculating normal mode vibrations. Daee, Lund, Plummer, Kassner and Hale (1972) succeeded in making detailed calculations on a wide range of cluster sizes using a newly developed method for calculating normal mode vibrations. Plummer and Hale (1972, 1973) have further developed and extended the theory to describe three dimensional clusters on idealized surfaces. The latter is applied to deposition nucleation. The essence of the method is applied to deposition nucleation. The essence of the method is that it parameterizes the hydrogen bond and treats it as if it were composed of a stretching harmonic oscillator and a bending harmonic oscillator. The basic

properties of the water molecule itself are introduced into the statistical mechanical equations in order to predict the features of the nucleation process.

Hagen and Kassner (1974) have generalized a technique due to Slater for estimating the frequency of bond breaking which could play an important role in estimating the magnitude of  $\Delta g_{SL}$  from first principles. At any rate these molecular dynamics techniques are an extremely powerful means of studying nucleation in hydrogen bonded systems and offer exciting possibilities for displacing the classical liquid drop model cited above. At this time better experimental information is badly needed in order to serve as a basis for developing the theory.

The major problem in attempting to investigate the homogeneous freezing of water droplets, is to devise a means of supporting the droplets for long periods of time without introducing contaminants so that the droplets can be slowly supercooled under the watchful eye of the investigator. Vonnegut (1948) studied the freezing of droplets placed on a polished chrome plated metal surface coated with polystyrene. The plate temperature was held constant and the freezing events recorded as a function of the time. The data was interpreted as a freezing probability per unit time. The freezing probability was higher to increase the degree of supercooling. Heverly (1949) studied drops suspended on small thermocouples. Hosler (1954) studied the freezing of droplets on a platinum surface and water sealed in small diameter Pyrex tubes which had undergone prolonged cleaning in chromic acid and rinsing with distilled water. Mossop (1955) indicated that Hosler's results must certainly have been affected by the film of water in contact with the inner wall of the tube. Bigg (1953) eliminated the solid surface support by suspending the droplets in the interface between two immiscible liquids of different density where they could not be contaminated by particulates in the surrounding air. Langham and Mason (1958) improved upon Bigg's technique slightly. Mossop (1955) found that the number of  $1\text{ }\mu\text{m}$  droplets which froze began to increase rapidly as the temperature fell below  $-40^{\circ}\text{C}$ . Mason (1956) formed droplets on ions in an aerosol free environment in a thermal diffusion chamber and by letting

the droplets fall through a thermal gradient. He found that essentially all of the droplets had frozen by the time  $-41^{\circ}\text{C}$  had been reached. Hoffer (1961) froze 60 to  $170\text{ }\mu\text{m}$  droplets suspended in silicone oil and observed median freezing temperatures in the range of  $-36$  to  $-370^{\circ}\text{C}$ .

The scatter in the data of different investigators would suggest that the individual experiments nearly always are affected by the mechanism of support. Perhaps some unique type of catalytic effect is afforded by each extraneous surface or source of contamination. While the data from a given experiment is much more likely to exhibit internal consistency, different experiments carried out under varying circumstances vary widely in their results. However, when all the data are plotted together there appears to be a reasonably well defined lower limit in temperature which can under certain circumstances be reached before nucleation sets in. It seems most likely that this limit represents the critical homogeneous freezing nucleation limit, i. e., the point at which the homogeneous freezing nucleation rate becomes sufficient to render the process observable. The present scatter in these nucleation results is such that the data are not very useful in developing the theory. The purpose of the zero-g experiment on homogeneous freezing nucleation is to remove as many of the experimental artifacts resulting from contamination as possible by making use of environment in which droplets can float freely in the chamber.

#### 1.3.4 Applications

Since nature frequently finds it difficult to supply adequate concentrations of ice nuclei, cloud droplets often supercool to  $-20^{\circ}\text{C}$  and occasionally to  $-35^{\circ}\text{C}$ . The coexistence of ice and supercooled water leads to riming which produces graupel which is one of the active mechanisms in the production of precipitation. It is this mechanism that one attempts to stimulate in cloud seeding operations. Development of cloud seeding techniques has proceeded for the most part on a trial and error basis with only a minimum understanding of the basic microphysical processes. These studies will enhance the understanding of those microphysical processes involved in the glaciation of cloud. Also in arctic cities such as Fairbanks, Alaska, ice fogs occur when temperatures fall below  $-35^{\circ}\text{C}$ .

Freezing studies such as this also have application in problems relating to the freezing of living cells. Here changes in the composition of the materials in the unfrozen and frozen material produces disruptive effects which are not always reversible. It is not understood how some living specimens can be frozen without permanent damage while others are damaged to such an extent that they are killed.

While the question of homogeneous nucleation is of less direct practical importance in itself, it does form the basis for understanding the more complex process of heterogeneous freezing nucleation which predominates in nature.

#### 1.3.5 Terrestrial Laboratory Limitations

Terrestrial laboratory experiments have been conducted to measure spontaneous freezing of pure water by several means. Droplets have been suspended by placing them on various surfaces, at the interface between two immiscible fluids of different density and by immersing the droplets in silicone oils of proper density. It is obvious from the data that the circumstances of many experiments provides some catalytic effects which is representative of that particular experiment. The catalytic effect is either due to impurities in the water or effects associated with the means of support.

Other experiments utilize droplets nucleated on ions in nuclei free air which are subjected to cooling while in free fall. In general these give most consistently to lowest obtainable temperatures. However, it should be recognized that droplets in free fall under the influence of gravity, frictional effects give rise to internal circulation, the effect of which on the freezing is unknown. It would be expected to have the most pronounced effect on larger droplets where the effect is more pronounced. Moreover, the estimate of temperature for droplets in free fall is more hazardous. Moreover, the freely falling droplet technique places limits on the range of cooling rates and drop sizes which can be studied in equipment of reasonable dimensions. One cannot place any confidence in the values of the surface free energy from different experiments where there is such a scatter in the data. What is needed is a single experimental technique which can yield freezing

nucleation data for an extremely wide range of drop sizes and for a wide range of cooling rates. Hopefully, such an experiment will yield sufficiently, consistent data so that one can have confidence in values of the surface free energy extracted from the data.

#### I. 3. 6 Zero-Gravity Opportunities

The primary restriction placed on the measurement of the homogeneous nucleation of ice in a terrestrial environment can be traced to the need to provide some type of support for the sample drop so that measurements of drop freezing temperatures can be made with sufficient accuracy and reproducibility to be useful. Support is also required to obtain sufficient ranges of drop sizes and cooling rates to verify the dependence of homogeneous nucleation on these parameters. The use of a zero-gravity environment permits the use of stationary free-floating sample drops and combines the advantages of the two terrestrial techniques (supported and free-falling drops) and avoids the disadvantages of each.

#### I. 3. 7 Quantification

Using present and foreseeable technology the zero-gravity environment represents the only means by which an experiment to make unambiguous measurements of the homogeneous nucleation of ice can be developed. The free-falling drop is the only terrestrial method that has any possibility, however, techniques for remote measurement of the drop freezing temperature with sufficient accuracy are not currently available nor does it appear likely that they will be developed in the near future.

#### I. 3. 8 Approach

##### I. 3. 8. 1 General

General comments on the operation of an expansion chamber system can be found under the Approach 1 of Class 6 - Scavenging.

While a study of the homogeneous nucleation of ice can be made using either an isometric or isobaric cooling of the chamber the long thermal time constant (approximately 150 seconds) of the chamber makes the use of an adiabatic

cooling preferable. Use of an adiabatic expansion causes the entire chamber to change temperature uniformly and the only thermal time constant to be considered is that of the drops themselves (thermal time constant in the range of 10-100 millisecc.). This will permit a much more accurate determination of drop temperatures than in either an isometric or isobaric cooling.

In the experiment, very pure water drops of the appropriate size and concentration are generated and placed in the chamber. The chamber and water drops are then cooled by an adiabatic expansion and the temperature at which the drops freeze is recorded using a time lapse camera.

The primary practical difficulties are the generation of "pure" water drops and maintaining the extremely cleanliness required. The presences of any ice nuclei either in the water drops or the chamber can alter the results by heterogeneous freezing.

One method of checking the cleanliness of the drop generation technique is to form the drops in the expansion chamber itself by homogeneous nucleation of drops from the vapor.

A second variation can be used to test for the presence of any memory effect associated with the homogeneous nucleation process. In this experiment, the chamber would start at a subsaturated condition and then form drops by homogeneous nucleation using an adiabatic expansion. The expansion would be continued until the drops have frozen. An adiabatic compression is then used to sublimate the ice crystals and then a second expansion to detect the presence of any memory effects. The initial relative humidity could be chosen to permit sublimation to occur at subfreezing temperatures preventing the ice crystals from melting.

Inclusion of these last modified experiments is strongly recommended to enhance the scope of the total experiment.



#### **I. 3. 8. 2 Experiment Parameters**

The important parameters along with the derived variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	6
Type	
Pollutant	
Pressure	
Temperature	
Relative humidity	
Charge	
Rate of cooling	2
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	4
Spin rate	

### 1.3.9 Procedure

To perform a homogeneous nucleation experiment in a terrestrial laboratory the expansion chamber must be increased in height to provide sufficient retention times as the drops fall due to gravitational settling. The increased size of the chamber will require longer purge times, faster cooling rates and larger quantities of purge gases than those required for a zero-gravity experiment.

The initial step is to turn on all the electrical components of the system and check to insure that all units are operational. The control and measuring circuits are then tested and any required calibrations performed. The entire system is then purged to remove all nuclei which might cause freezing if they come in contact with a supercooled drop. The purge gas should be the same noncondensable gas as that planned for use in the experiment. While the system is being purged the control program for the particular experiment planned is loaded into the control computer.

Once the system has been adequately purged the expansion chamber ports are closed and the gas flow to both the humidifier and drop generator adjusted to predetermined values. Thermal control of the expansion chamber is initiated to bring the chamber to the correct starting temperature. Thermal control of the humidifier and conditioning chamber is also turned on. Once the humidifier and conditioning chamber temperatures have stabilized at the correct values, the drop generator is turned on and the drop size and concentration of the sample monitored as it leaves the conditioning chamber.

Monitoring of the sample for drop size and concentration continues until a stable condition is reached and the test sample has the required drop size and concentration. The ports of the expansion chamber are then opened and the sample flushed through the chamber. Monitoring is transferred to the outlet of the expansion chamber and flushing continues until the drop size and concentration become stable with respect to time. The expansion chamber ports are then closed and the pressure control activated to maintain the initial chamber pressure constant while the chamber comes to thermal equilibrium.

While the expansion chamber is coming to thermal equilibrium the drop generator is turned off and the system purged with clean gas to remove any remaining drops. The purge gas is then turned off.

After the expansion chamber has come to thermal equilibrium the cooling/expansion cycle is initiated. During the expansion the drops are photographed using the time lapse camera. The pressure, chamber wall temperature, calculated gas temperature, liquid water content, and drop size are also recorded.

As knowledge of the temperature at which drop freezing begins becomes available the initial portion of the cooling/expansion cycle can be speeded up to shorten the time required to reach the temperatures of interest. This will permit the use of lower cooling rates at the lower temperatures without increasing the duration of the experiment.

When all the drops have frozen the chamber is heated and compressed until the ice crystals have melted, then cooled and expanded to refreeze the drops. The refreezing is used to investigate any possible memory effects.

At the completion of the experiment the ports of the expansion chamber are opened and the chamber purged with clean gas to remove the remaining water drops and ice crystals. Purging is carried out at a temperature above  $0^{\circ}\text{C}$  to remove any ice from the chamber walls.

While the chamber is being purged the time lapse camera is unloaded and the film stored for processing. The computer data is checked for any major malfunctions and then transferred to permanent storage. When the chamber has been purged the ports are released, the purge gas turned off and the system shut down. (See Figure I-3.)

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on all electrical components	5
• Check operational readiness of components	5
• Check and calibrate control and measuring circuits	
• Turn on noncondensable gas supply and sump pump	0.5
• Open expansion chamber ports	0.5
• Purge system	10
• Load control program into computer	(5)
• Load time-lapse camera	(3)
• Close expansion chamber ports	0.5
• Turn on expansion chamber thermal control	
• Turn on humidifier thermal control	0.5
• Turn on conditioning chamber thermal control	
• Adjust gas flow to humidifier	0.5
• Adjust gas flow to drop generator	0.5
• Turn on drop generator	1
• Turn on drop size spectrometer and mass meter	1
• Turn on hygrometer	0.5
• Monitor drop size, mass and concentration and sample relative humidity	10
• Adjust humidifier temperature	
• Adjust drop generator	
• Open expansion chamber ports	0.5
• Transfer monitoring point	0.5
• Monitor drop size, mass and concentration	8
• Close expansion chamber ports	0.5

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on expansion mechanism, pressure control, laser, liquid water meter, drop size meter, video, gas temperature	1
• Let chamber come to thermal equilibrium	10
• Turn off drop generator, humidifier, conditioning chamber, thermal controls, drop size, mass and concentration meter, and hygrometer	(0.5)
• Purge system	(4)
• Turn off noncondensable gas supply and sump pump	(0.1)
• Turn on time lapse camera	0.5
• Start cooling/expansion program	30
• Stop time-lapse camera	0.5
• Turn off pressure control, expansion mechanism, laser, liquid water meter, drop size meter, video and gas temperature	1
• Heat chamber to +10°C and turn off expansion chamber thermal control	5
• Open expansion chamber ports	5
• Turn on noncondensable gas supply and sump pump	0.5
• Purge chamber	8
• Unload time-lapse camera and store film	(3)
• Check computer data	(4)
• Transfer computer data to permanent storage	(1)
• Close expansion chamber ports	0.5
• Turn off noncondensable gas supply and sump pump	0.5
• Shut down system	10

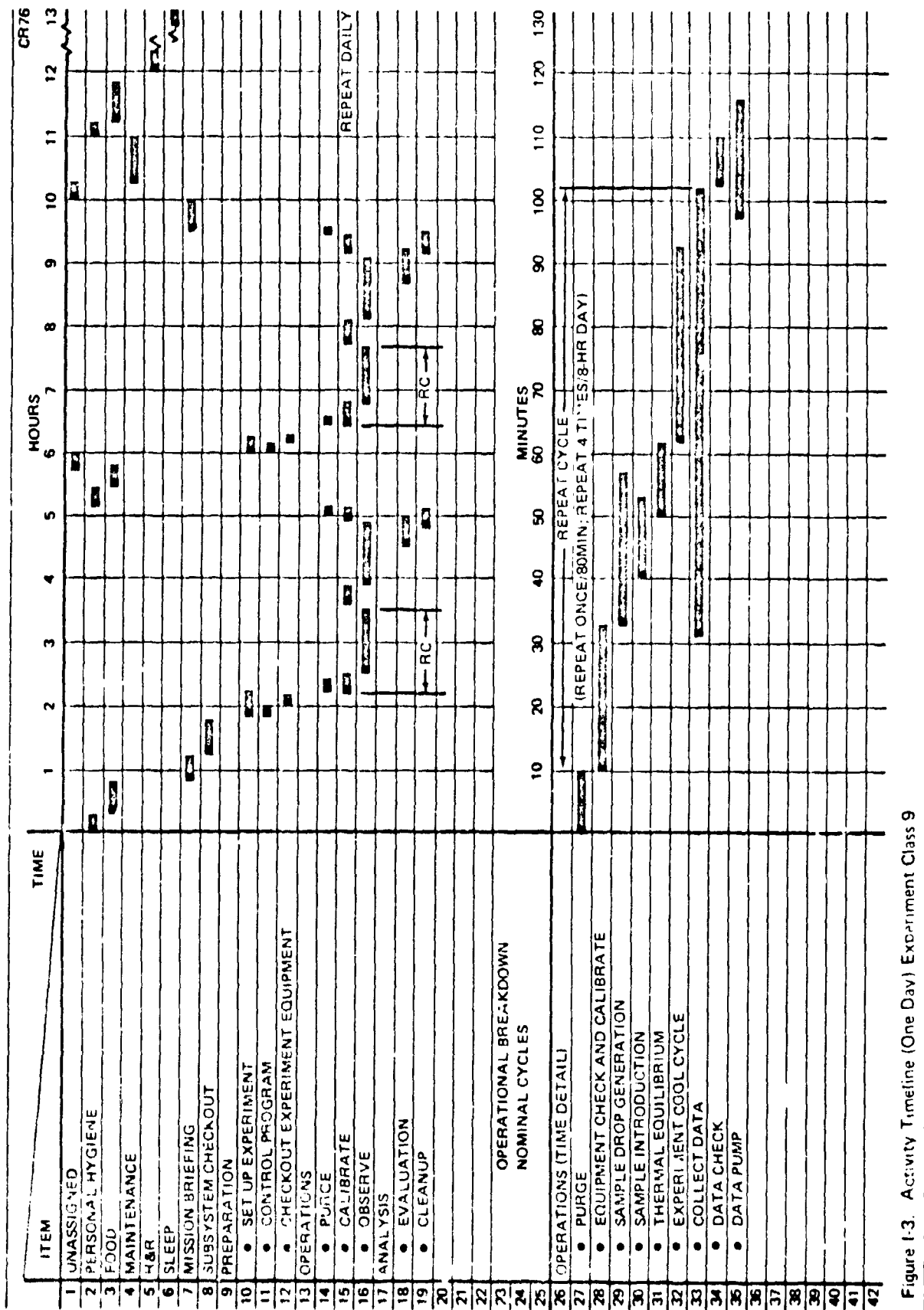


Figure 1-3. Activity Timeline (One Day) Experiment Class 9

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Appendix J  
CLASS 10  
COLLISION-INDUCED FREEZING

J.1 INTRODUCTION

Many observations in natural clouds have shown that the concentrations of ice particles are sometimes much greater than the measured concentrations of ice nuclei effects at the cloud top temperature, Koenig (1), Braham (2), Mossop et al (3), Koenig (4), Hobbs (5) and Mossop (6, 7).

Laboratory experiments have suggested several ways in which the number of ice particles can be multiplied in a cloud:

- A. ice splinters formed at the surface of a frozen drop, Findensein (8), Schaefer (9) and Muchnick (10).
- B. fragmentation of freezing water drops, Langham-Mason (11), Mason-Maybank (12), Evans-Hutchinson (13), Scott-Hutchinson (14), Kuhns (15), Hobbs-Alkezweeny (16) and Johnson-Hallet (17).
- C. collision of supercooled drops and an ice crystal, Brownscombe-Hallet (18).
- D. fragmentation of ice crystals by collision with supercooled droplets, Hobbs-Farber (19) and Ono (20).
- E. collision induced freezing, Podzimek (21, 22), Hobbs (23), and Alkezweeny (24).

Of all the mechanisms the last one is the less known and understood and because it appears to be independent of the ice nuclei it could result in a supplementary source of ice particles in a cloud.

A better knowledge of this mechanism will help cloud modeling and may open new ways for weather modification.



## J.2 OBJECTIVE

The objective of the experiment is to determine the efficiency of the collision induced freezing process and how different external conditions can influence it.

## J.3 SCIENTIFIC JUSTIFICATION

The formation and propagation of the ice phase in a cloud affects its dynamics and its collidal stability. Any improvement in the knowledge on the ways ice particles are produced will help our understanding of the clouds evolution.

Besides the fact that collision induced freezing is accomplished with a reduction in the free energy of the system, there is not enough information to support any interpretation of why the collision can trigger the freezing.

The observations available show that the collision of drops of different sizes can trigger the freezing, Alkezweeny (24), and also that drops of similar sizes when brought together may produce an ice particle, Podzimek (22).

Influence of the temperature has been reported by Podzimek (22). All the information points to the existence of a freezing mechanism but there is not enough evidence to understand it.

To evaluate the potential of this mechanism, a better knowledge about the influence of drop sizes, temperature, humidity, turbulence, electric and acoustic fields is highly desirable. It would also be important to determine the dependence on the ice nuclei.

## J.4 APPLICATIONS

A better knowledge of the collision induced freezing nuclei find application in the following areas.

### J.4.1 Cloud Physics

Ice phase generation and propagation and its influence in the cloud dynamics and colloidal stability.

#### **J. 4. 2 Weather Modification**

The evaluation for cloud seeding potential is based largely on ice nuclei concentrations. The possible existence of an independent source of ice particles may change the evaluation patterns.

If the collision induced freezing proves to be independent of the ice nuclei, then a new way to modify clouds would be possible.

#### **J. 5 TERRESTRIAL LABORATORY LIMITATIONS**

All the information gathered has been obtained with

- A. drops hanging from wires
- B. small cloud chambers
- C. cloud particles collected in an airborne formvar replicator.

The presence of the support introduces an unknown amount of distortion and there is no way to estimate its magnitude nor its direction.

The cloud diameter and the particles collected in a cloud give only overall results, thus making it very difficult to discern the separate influences of different parameters.

#### **J. 6 ZERO-GRAVITY OPPORTUNITIES**

The absence of gravity will allow the elimination of the need of support and to study the individual behavior of pairs of drops. The influence of external conditions can be studied in detail along with that of drop size. One particular advantage will be to determine the production of secondary ice particles through splinter formation and drop fragmentation.

The experiments in zero gravity conditions may be the only way to get an accurate and complete understanding of this complex process.

#### **J. 7 PROCEDURE**

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to the low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure J-1).

### Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	10
Type	
Pollutant	
Pressure	3
Temperature	4
Relative humidity	4
Charge	
Rate of cooling	
Time	
Sound	3
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	3
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Stabilize the SDI for the particular temperature range	15
● Inject large drops	1
● Inject second drop	1
● When second drop is injected, start taking pictures and continue to do so during the next 5 minutes taking a picture every 10 seconds.	5
● Remove all the particles in the chamber	3
● Repeat	

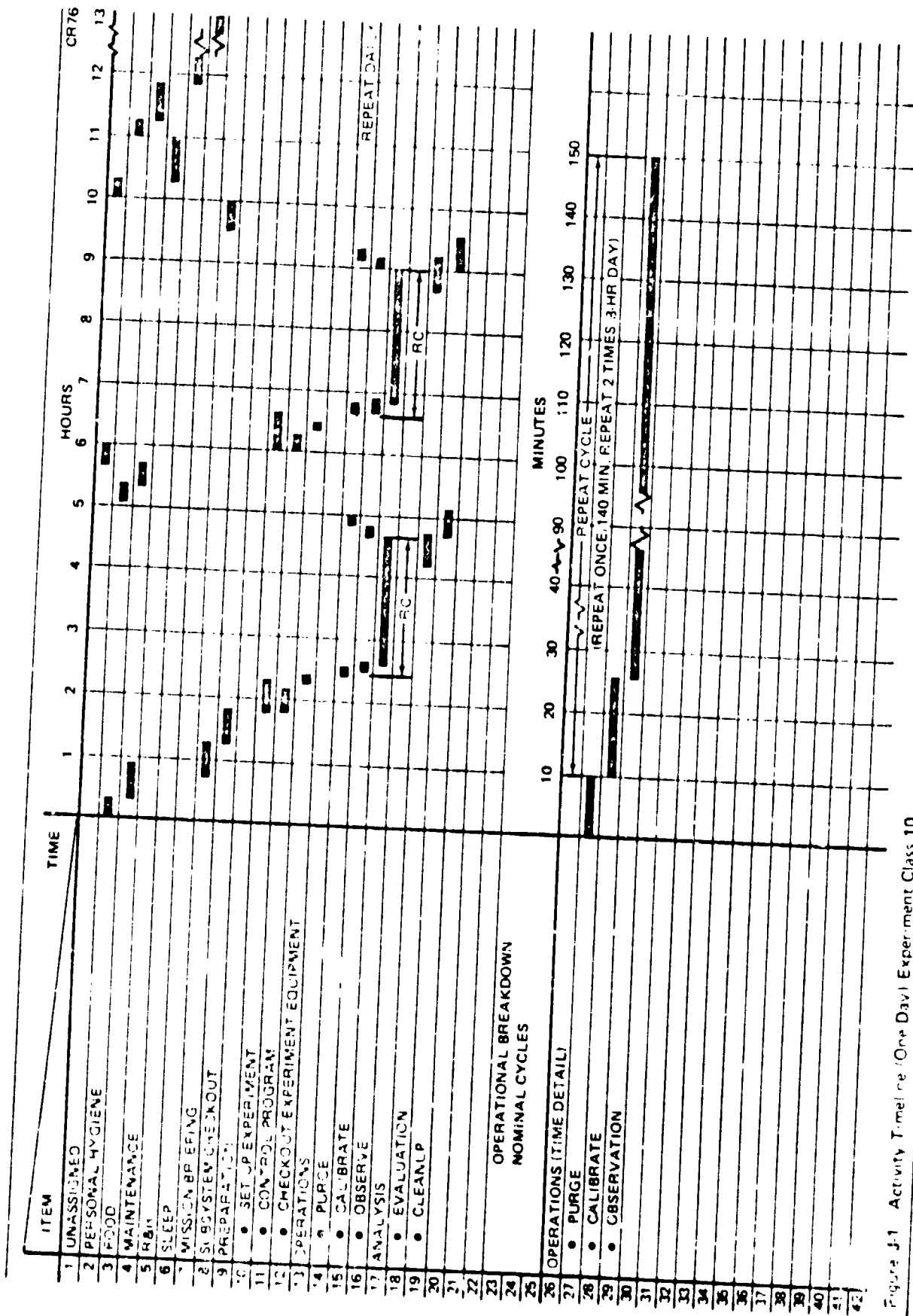


Figure J-1 Activity Timeline (One Day) Experiment Class 10

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Appendix K  
CLASS 11  
SATURATION VAPOR PRESSURE  
(SUPERCOOLED WATER)

K.1 SATURATION VAPOR PRESSURE APPROACH 1

K.1.1 Introduction

The formation and growth of ice phase in supercooled clouds are important processes in understanding the evolution of the clouds and in modifying them. These processes during the phase change from the supercooled liquid directly to solid or indirectly via gaseous phase, proceed according to the free energy difference between them. The free energy difference is a function of vapor pressure difference between the two phases. Since ice is stable under temperatures lower than 0°C, the saturation vapor pressure has been determined in sufficient accuracy, whereas the direct vapor pressure measurement of supercooled water has not been carried out to very low temperatures, approaching -40°C where duration of the supercooled condition of water becomes extremely short due to the homogeneous nucleation of ice in it.

When the vapor pressure measurement of supercooled water is carried out using a container, the supercooled condition does not last long due to the influence from the container wall. If one attempts to directly measure the vapor pressure using an aerosol form of water, the low vapor pressure at the temperatures in question, the Kelvin effect, particle settling, and ice nucleation will interfere with the measurement. However, the unique low gravity condition in the space laboratory provides possibilities of easing some of the restrictions experienced in the terrestrial laboratories.



#### K.1.2 Objective

It is the objective of this experimental study to accurately determine the saturated vapor pressure of supercooled water down to a temperature as close as possible to the homogeneous nucleation point of ice utilizing the special advantage of the Zero Gravity Laboratory.

#### K.1.3 Scientific Justification

The phase transition of supercooled water into ice is of great importance in cloud physics and weather modification. The free energy difference involved in the phase transition is the driving force, and it is a function of saturated vapor pressures of ice and water. Since ice is stable at temperatures below 0°C under pressures of atmospheric phenomena, the vapor pressure data are obtained in sufficient accuracy. On the other hand, due to the difficulty of holding the supercooled state, reliable data for the saturated vapor pressure over supercooled water are unavailable at low temperatures. Therefore, if the unique low gravity condition of the space laboratory permits accurate determination of the vapor pressure, the data obtained will be an important addition to the fields of cloud physics, weather modification, and atmospheric sciences.

#### K.1.4 Applications

Since it is a set of data that is to be determined in this study, the possibility of its application spans from the fundamental field to applied subjects concerning such as the growth and evaporation of supercooled water droplets, the diffusional ice crystal growth kinetics in the supercooled clouds and fogs, and the thermodynamics of supercooled cloud glaciation including changes in cloud buoyancy and temperature.

#### K.1.5 Terrestrial Laboratory Limitations

The terrestrial laboratory limitation of this research subject originates from the difficulty of keeping the supercooled water in a vessel, and this naturally leads one to consider utilization of the wall-free condition of aerosol water or fog. However, this approach introduces a dilemma with regard to the

gravitational settling and the Kelvin effect. That is, small particles minimize the settling problem but the Kelvin effect which increases the equilibrium vapor pressure due to the surface energy contribution in combination with the curvature of the surface, shifts the value of the vapor pressure away from that of the flat surface. The saturated vapor pressure of supercooled water over the flat surface is the main value to be determined in this study.

#### K. 1. 6 Zero-Gravity Opportunities

The extremely low gravity condition of the space laboratory permits one to solve the dilemma mentioned just above. Both the particle falling problem and the Kelvin effect can be avoided by suspending sufficiently large droplets in the space laboratory.

#### K. 1. 7 Approach

##### K. 1. 7. 1 General

The principal of this study is first to grow water droplets in clean nuclei free air starting from homogeneously nucleated droplets in the SDI being operated at temperatures above 0°C. Then, the median plane temperature of the chamber,  $T_M$ , is lowered to a desired level while maintaining the water saturation condition in the plane by controlling the temperature difference between the top and bottom plate,  $\Delta T$ . Observing the backward laser beam interference from a droplet staying in the median plane, the condition of showing no movement of the interference rings will be determined in terms of  $\Delta T$  and  $T_M$ . Under that condition, the water vapor pressure in the median plane is exactly at water saturation. From  $\Delta T$ ,  $T_M$  and available data of ice vapor pressure, the saturation vapor pressure of the supercooled water can be estimated.

The experimental procedure is as follows. The SDI will be used under low pressure. The low pressure operation will allow the effect of top and bottom plate conditions to swiftly reach the median plane.

After purging the chamber with nuclei free air under the room temperature and pressure, an adiabatic expansion will be made with small  $\Delta T$  above  $0^{\circ}\text{C}$  so that homogeneous nucleation of droplets takes place. Then, the pressure will be brought back slowly to 1 atm, and the number concentration of fog droplets will be reduced to the desired level by diluting it with nuclei free air. The chamber pressure will be reduced again and the fog droplets will be exposed to a supersaturation by increasing  $\Delta T$ . When the size of the fog droplets in the median plane of the chamber reaches a predetermined value, temperatures of top plate ( $T_1$ ) and bottom plate ( $T_2$ ) will be lowered while sustaining the water saturation condition in the plane, and  $T_M$  will reach the temperature of measurement. One of the droplets on the median plane will be illuminated by a laser beam and the interference of the backward reflected lights from two surfaces of the droplet in the center will be observed through a microscope of long working distance. The number of interferences observed in a unit time as repeating cycles of dark and bright conditions in the center of the droplet will be recorded together with  $T_1$ ,  $T_2$ ,  $T_M$  under a supersaturated condition. If the supersaturation is large, the frequency of the cycle is high and  $\Delta T$  has to be reduced without changing  $T_M$ .

By reducing  $\Delta T$  in this manner, the condition where the frequency of the interference cycle becomes zero will be sought, and when the condition is achieved,  $T_1$ ,  $T_2$ , and  $T_M$  will be recorded. Then,  $\Delta T$  will be further reduced so that the median plane condition becomes undersaturated with respect to water. From this point,  $\Delta T$  will slowly be increased to find the same condition of frequency zero, and under the condition,  $T_1$ ,  $T_2$ , and  $T_M$  will be recorded.

The same procedure will be repeated for different temperatures. The droplets grown in this manner are extremely clean and should stand supercooling for a long time.

#### K.1.7.2 Instrumentation

The required instrumentation includes an ordinary supporting facility to produce clean filtered air, a modified SDI for expansion operation, a laser device, and a microscope of very long working distance.

### K.1.7.3 Experiment Parameters

The important parameters along with the desired variations for each parameter are given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	1
Type	
Pollutant	
Pressure	1
Temperature	15
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### K.1.8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to the low-g environment. Additional effort will be required to make these timelines operational effective and efficient (see Figure K-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Purge SDI Chamber	5
● Open $\Delta T$ with TM at room temperature	2
● Expand chamber air for homogeneous nucleation with small $\Delta T$ .	1
● Bring pressure back slowly to 1 atm.	3
● Dilute fog.	3
● Lower chamber pressure	3
● Resume $\Delta T$ , grow fog droplets	10
● Lower TM to measurement temperature while maintaining water saturation in median plane.	10
● Activate laser.	2
● Find interference cycle zero point	25
● Record $T_1$ , $T_2$ , TM	
● Recycle at other temperature	

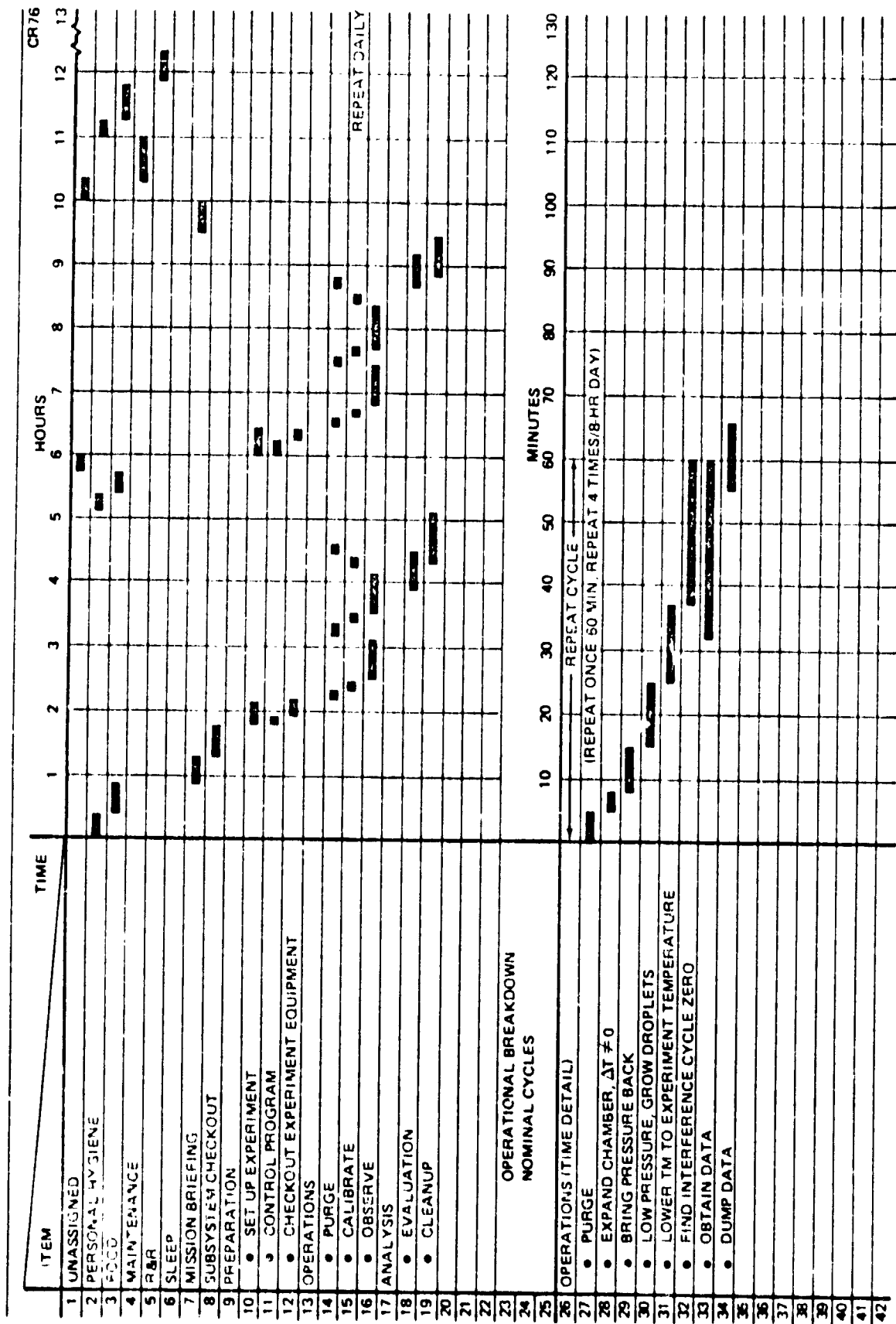


Figure K-1. Activity Timeline (One Day) Experiment Class 11

## K.2 SATURATION VAPOR PRESSURE (SUPERCOOLED WATER) APPROACH 2

### K.2.1 Introduction

The measurement of the saturation vapor pressure over supercooled water has been accomplished with good results (see Dorsey, 1940) to temperatures of -5°C. This experimental work was conducted prior to 1940 as were measurements on the vapor pressure over ice.

The saturation vapor pressure is given by the Clausius-Claperyon Equation (see Holmboe, 1945).

$$\frac{1}{e_s} \frac{de_s}{dT} = \frac{L}{R_w T^2} \quad (1)$$

where  $e_s$  is the saturation vapor pressure,  $L$  is the latent heat of vapor to liquid,  $R_w$  is the gas constant for water vapor and  $T$  is the temperature.

A comparison among the experimental results for vapor pressure above 0°C, a water to vapor system shows that the error from the simple Clausius-Claperyon to the real gas behavior, which is not ideal, shows errors in the less than 0.4 percent range (c.f. Table K-1). Certainly such errors are not of great significance in the growth equation (Byers, 1965) which, in slightly different forms, governs both ice and water droplet growth.

Table K-1  
SATURATION VAPOR FROM DIFFERENT EQUATIONS

-20°C	-10°C	0°C	+10°C	+20°C	
0.1273	.2873	.611	1.232	2.37	Clausius-Claperyon
0.1254	.2862	.611	1.228	2.339	Magnus
.12540	.28627	.61078	1.2272	2.3373	Goff-Gratch



It is

$$r^2 = r_o^2 + (S - 1) (1 + 0.22 F R_e^{1/2}) \Delta t / (A + B) \quad (2)$$

where  $R_e$  is the Reynolds number,  $F$  is the ventilation coefficient,  $r$  is the radius of the droplet,  $S$  is the ratio of the vapor pressure to the saturation vapor pressure in the environment,  $A$  and  $B$  are functions of temperature and pressure. It is apparent that one percent change in supersaturation is not significant

The supersaturation is greatly affected by the change in concentration of a solute within a droplet. This is given by Raoult's (1882) Law as

$$\frac{e}{e_s} = 1 - \frac{\bar{Z} M}{\bar{Z} M + M_o} \quad (3)$$

where  $\bar{Z}$  is the Van't Hoff factor,  $M$  is the number of moles of solute and  $M_o$  is the number of moles of solvent. Close to a saturated solution, the Van't Hoff factor varies, at least for NaCl, widely (McDonald, 1953). This behavior should be investigated.

There is no fundamental reason to doubt the theory as given by (1). However, experimental verification should be highly important since the diffusional growth of ice crystals is maximal between -10 and -20°C. The growth of ice crystals is fundamental to weather modification.

#### K.2.2 Objective

The purpose of these experiments is to measure the saturation vapor pressure at temperatures below 0°C for "pure water" and for several salt solutions. The salt solutions would include: NaCl,  $(NH_4)_2SO_4$ ,  $Na_2SO_4$ .

### **K.2.3 Scientific Justification**

The results of these experiments will:

- a) Fix the supersaturation ratio (water/ice) for ice crystal growth  
 $T > -20^{\circ}\text{C}$ .
- b) Determine the behavior of the Van't Hoff factor for certain salts  
(this is important in the initial growth process and in the final stages of evaporation).

### **K.2.4 Applications**

- a) Weather modification. The diffusional growth of ice is calculated in targeting precipitation. Experimental verification of the vapor pressure puts the calculations on a firmer basis.
- b) Calculations of diffusional drop growth below  $0^{\circ}\text{C}$ .
- c) Condensation and evaporation on salt nuclei below  $0^{\circ}\text{C}$ .

### **K.2.5 Terrestrial Laboratories' Limitations**

Terrestrial laboratories are limited since large, the order of  $200\text{ }\mu\text{m}$ , drops are the most useful for experimentation. This means that the drops must be supported in some manner because the experiment will take several hours to perform. Even though extremely fine plastic or spider web strands can be used to support droplets, the experiment is not the same as with an unsupported droplet. A supported droplet may be nucleated by the fiber, thereby increasing the probability that a particular droplet will freeze. The positioning and supports are serious complicating factors.

Measuring the vapor pressure over bulk water is impractical due to the higher probability of freezing and the longer time constant necessary to reach equilibrium.

### **K.2.6 Zero-Gravity Opportunities**

The crucial question lies in the heterogeneous nucleation rate of spider webs and plastic strands. If any nucleation takes place at  $-20^{\circ}\text{C}$  from the support source then the experiment becomes intractable on the Earth and the only possibility of obtaining the data lies in the zero gravity experiment. This is especially true since  $200\text{ }\mu\text{m}$  diameter droplets have, on Earth, a fall velocity greater than a meter/sec and must be supported.

Under conditions of near zero gravity with sufficiently pure water, the drops should only freeze homogeneously; thus even over time periods of several hours, no drops should freeze at  $-30^{\circ}\text{C}$ .

The differential of success in a terrestrial laboratory or at near zero  $g$  is heterogeneous nucleation. The probability of a drop freezing increases with total number of drops (volume) and cannot be quantified.

#### K. 2. 7 Approach

##### K. 2. 7. 1 General

The experiment is difficult to perform and a limited amount of terrestrial experimentation above  $0^{\circ}\text{C}$  should attempt to duplicate the better experimental results. Fundamentally, the difficulty arises from the long time steps necessary to approach equilibrium (some 500 seconds even with close drop spacing). If the experiment is conducted under steady temperature decrease to alleviate this problem and equilibrium is barely approached, the accuracy of measurement becomes the principal problem.

The saturation vapor pressure over a droplet is a function of temperature only. The determination of droplet temperature is a primary measurement. The diffusion of the vapor from the drop is pressure dependent and follows equation (2). The rate of mass change from a droplet depends on the rate at which the diffusion occurs and heat is supplied to the droplet.

Terrestrial experiments, due to the difficulties in supporting the drops, generally only grossly approach equilibrium. If the measurement is to be excellent, a good approach to equilibrium is needed.

The experiment must be performed in a chamber with non-nucleating walls, presumably the general chamber.

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It is of interest to note that homogeneous nucleation can be studied with the same droplet set simply by continued decrease in temperature.

The parameters of:

drop temperature;

temperature;

absolute pressure; and

differential pressure

must be measured accurately. The differential pressure to  $10^{-3}$  dynes  $\text{cm}^{-2}$ ; the absolute to .1 dyne  $\text{cm}^{-2}$ .

The temperature of the drop and the temperature of the chamber to within .01 °K.

The temperature and the drop temperature should be within .01 °K to assure equilibrium.

The parameters of:

temperature and

water purity

must be controlled.

Observations must be made to insure that no drops have frozen. (This requirement is not unique to this experiment. It also pertains to all experiments that look at drop freezing.)

Drops of pure water must be generated and positioned. A drop generator of the type described by Abbott and Cannon (1972) appears very promising for this type of study. This generator permits generation of single droplets at a fixed rate or on demand. This feature minimizes the total amount of pure water needed to perform this experiment.

Drop positioning using acoustical or other techniques must be developed.

It is difficult to assess how to approach the experimental problem other than conceptually since all the reported efforts to supercool bulk water have failed and few others have been attempted.

The experiment, performed in a terrestrial laboratory, would require:

- a) Non-nucleating chamber walls;
- b) Non-nucleating supports for droplets;
- c) Measurement of drop temperature;
- d) Measurement of air temperature;
- e) Measurement of total pressure;
- f) Measurement of vapor pressure;
- g) A means of drop generation;
- h) A scanning system for ice or frozen drops.

The items which would be different in zero g are:

- a) Item b), a source of probable trouble, would be unnecessary;
- b) The drop generation technique;
- c) Droplet placement.

The experimental trade-offs are:

Number of drops evaporating which governs the time to reach diffusive equilibrium. To illustrate this point, consider a single droplet in a chamber. With evaporation, the concentration at a particular point will be dependent upon the diffusion coefficient which is a function of time. If, however, the vapor is diffused into overlapping volumes, then the time to approach equilibrium decreases. Thus the use of many drops decreases the time necessary to complete the experiment. However, as the volume of water increases so does the probability that a drop will freeze since the nucleation rate is a function of volume and time.

The experiment must be performed in a manner to maximize diffusion, and to minimize time and thereby freezing of droplets by homogeneous nucleation. For instance, at a drop separation of 1 cm the time required to approach equilibrium is some 500 seconds. Now in the general chamber there would have to be some 27,000 droplets of 100  $\mu\text{m}$  in diameter. These droplets would have to be positioned in some manner to avoid gross imbalance in the diffusion field. This positioning question is probably the most difficult part of the experiment to perform.

The chamber must be made of a non-nucleating surface. Prior experience leads to the use of highly polished stainless steel or teflon.

An experimental trade-off can be made at this point; fewer droplets, say, the order of a hundred of 200  $\mu\text{m}$  could be used. The time to reach equilibrium becomes, however, excessive (greater than 2000 sec.).

The general purpose chamber in this experiment must be temperature controlled, but not to an absolute temperature. The decrease of temperature must be controllable, based on the  $\Delta(T - T_{\text{drop}})$ . A sensitive proportional controller can be used to decrease the temperature towards equilibrium. The temperature range of primary interest is 0 to  $-25^{\circ}\text{C}$ . (It is herein assumed that the chamber is cooled with thermoelectric units or heat pipes and that sufficient insulation is provided to have a uniform temperature at the chamber wall.)

The experimental approach is to:

- a) Lower the temperature with an initially saturated chamber at  $0^{\circ}\text{C}$ . The temperature lowering means that the drops will grow. This procedure tends to prevent water or ice forming on the chamber wall.
- b) Lower an increment ( $2^{\circ}\text{C}$ ) in temperature and then stabilize the temperature.
- c) Check for freezes.
- d) Lower to  $-30^{\circ}\text{C}$  by  $2^{\circ}\text{C}$  steps or until a freeze occurs.
- e) Repeat for the saturated salt solution droplets.

#### K.2.7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size nuclei	
Size-droplet	1
Type	4
Pollutant	
Pressure	

<u>Parameters</u>	<u>Variations</u>
Temperature	20
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### K.2.8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to the low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure K-2).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Purge chamber	5 mins
• Lower pressure to 50 mb.	1 min
• Humidify	10 mins
• Generate droplets	51 mins
• Close off differential pressure	10 sec
• Decrease temperature 2°C and equilibrate	30 min
• Scan for frozen droplets	30 sec
• Repeat 15 times (-30°C)	457.5 min total
• Repeat all steps 10 times for pure water and 5 times for each salt solution	
<div> <div>Total Time</div> <div>Pure Water</div> <div>4786</div> <div>minutes</div> <div>79.8 hrs</div> </div> <div> <div>Salt Solutions</div> <div>7179</div> <div>minutes</div> <div>119.65 hrs</div> </div>	



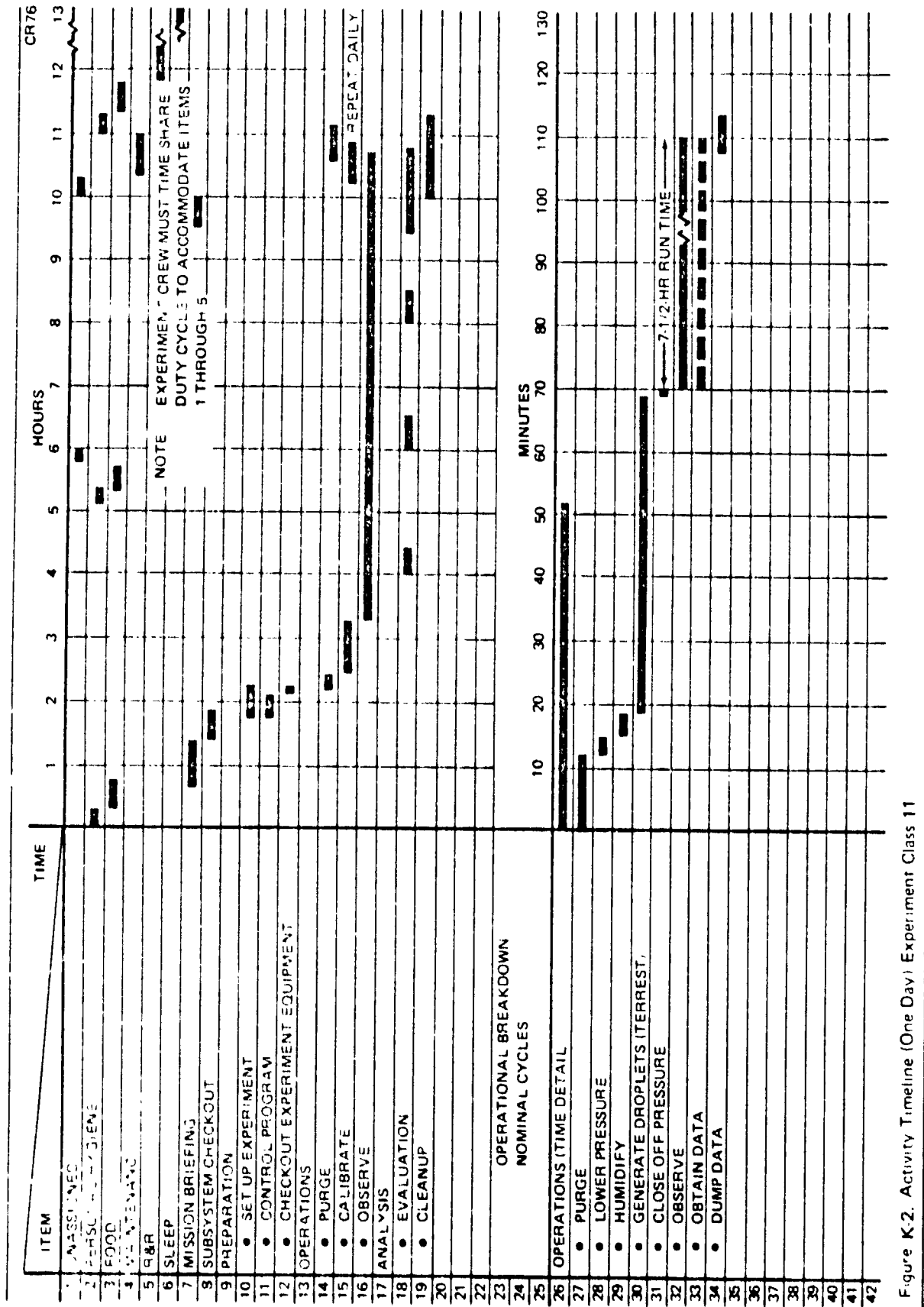


Figure K.2. Activity Timeline (One Day) Experiment Class 11

The difference between terrestrial and zero g are two-fold.

- a) The drops would not be supported on strands in zero g but positioned acoustically, and,
- b) The time for each step would be reduced to approach equilibrium within 2 percent (approximately 10 minutes).

#### K.2.8.1 General Information

##### Purging

Chamber to be purged before each data run.

##### Cleanliness

The chamber is not to contain any nucleating particulates or any gases other than  $H_2O$  vapor and  $N_2$ . Chamber pressure is to be 50 millibars (at  $0^\circ C$ ) so that diffusion is maximum, yet no radiational cooling droplets to wall.

##### Control Methods

$\Delta T/\Delta t$  is to be proportionally controlled from quartz thermometer close to the wall and  $T_{drop}$ . Rapid cooling towards the  $2^\circ C$  step slowing as the  $T_{drop}$  approaches the set temperature.

Drop Generation (drops - detailed elsewhere). Drops of salt solution will be 100 percent saturated with respect to the solute at  $0^\circ C$ .

##### Control

Position - unspecified technique - acoustic offers greatest promise.

The water must be pure. See Haller and Duecker (1960). If small enough amounts are needed, then making it from  $O_2$  and  $H_2$  may prove the best technique. Certainly the pure water must be made in a terrestrial laboratory. Standard purification techniques would probably provide pure enough water.

#### K.2.8.2 Conclusions

The experiment is difficult and three critical problems must be solved if it is to be successful. The solution of the three problems of drop scanning for freezes, drop temperature and drop positioning will also be of importance to other experimental efforts such as homogeneous nucleation.

The complexity and length of time necessary to perform the experiment, dictates that this experiment be performed after the majority.

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Appendix L  
CLASS 12  
ADIABATIC CLOUD EXPANSION

L.1 INTRODUCTION

Until recently, attempts to simulate the adiabatic expansion of a parcel of air in a convective cloud have met with, at best, only limited success. Gunn's attempts to utilize an abandoned mine shaft (Gunn-1952, Gunn and Allee-1954) and a 3,000 m<sup>3</sup> Horton sphere (Gunn and Allee-1954, Gunn-1958) are two of the better known examples where the technology of the day was just not adequate to the requirements of the task. Evans' simulation chamber (Edwards and Evans-1961) while experiencing some success did not possess the flexibility of operating parameters required for a comprehensive study of cloud physics problems. Podzimek (1964), using a chamber originally designed and built by Findeisen (1939), was able to simulate updraft velocities in the range 2 m/sec to 60 m/sec. However, lack of an adequate technological base at that time and lack of cloud chamber flexibility again limited the range of experiments. A major difficulty, which has plagued past efforts, has been the lack of remote sensing techniques permitting measurements of the experimental parameters within the chamber without introducing the disturbing influence of sensor probes. It is only relatively recently that such remote sensing techniques have been successfully developed.

Efforts underway in the U.S.S.R. (Volkovitskii-1965, Kartsivadze et al. - 1974, Laktinov and Volkovitskii-1969) have been directed toward the construction of extremely large chambers in attempts to increase the cloud retention time and reduce the reheating of the gas due to the walls by reducing the surface to volume ratio. The use of large chambers creates substantial problems in the development of sensing techniques, maintenance of uniformity throughout the chamber, production of homogeneous nuclei samples and suppression of convection currents within the chamber. Such giant chambers are

useful for calibrating transmissometers and a number of other experiments. However, they have not proven to be very successful for studying basic cloud formation processes.

## L.2 OBJECTIVE

Simulation of the conditions of the early stages of the life cycle of a parcel of air involved in an atmospheric cloud formation process utilizing time scales comparable to those observed in natural cloud and fog formation.

## L.3 SCIENTIFIC JUSTIFICATION

The whole concept of weather modification is predicated not only on the supposition that nature does not always provide an atmospheric aerosol conducive to the development of the most desirable type of precipitation and that man can alter natural processes in a predictable way by artificial seeding.

The general approach of simply matching specific causes with observed effects, without having to resort to an understanding of the intervening processes binding to a specific effect, have produced limited gains. The fact remains of course that in cloud physics, the most promising mode of intervention appears to be that which affects the phase transition of water from its vapor to liquid (or solid phase); such intervention clearly is concerned with nucleation, activation, and growth (or evaporation) processes, and is therefore tied into the introduction of artificial nuclei. Unfortunately, the effects of nuclei and their interaction with the dynamics being not too well understood, the consequences of the introduction of seed nuclei into a cloud process is not always predicable (Warner-1973, Neyman and Scott -1973).

The experimental approach used in the past has been to measure the nucleation and activation efficiencies of different types of nuclei using various techniques (fast expansion chambers, isothermal freezing chambers, etc.) none of which actually duplicate conditions in a natural cloud. Separate experiments have been carried out to measure the diffusional and coalescence growth rates of cloud droplets, again under conditions which do not truly simulate natural clouds. A well-known complexity consists of the situation

in which the processes of activation of nuclei and growth in natural clouds are occurring simultaneously and in competition with each other for the available water vapor being released by the driving thermodynamic processes. In terrestrial chambers, this already complex situation is further complicated by collision coalescences induced by the differential fall velocities. Before the microphysics can be effectively modeled, it will be necessary to investigate through a series of comprehensive simulation experiments the conditions existing in natural clouds such that a deeper understanding of the entire precipitation process can be achieved. The opportunity of utilizing both the terrestrial and the zero-g environment to study both isolated and simultaneously occurring processes offers an unusual opportunity to unravel some of the complexities and to develop a more precise parameterization for composite microphysical models.

Due to the large scale effects in the formation and development of clouds which can be caused by relatively small changes in the nuclei characteristics, an adequate understanding of the formation and development of clouds on well characterized aerosol spectra is necessary before a systematic program can be developed to either suppress inadvertent weather modification due to air pollution products or produce effective deliberate weather modification through artificial seeding techniques.

#### L.4 APPLICATIONS

The increased knowledge gained through a study of the complete precipitation process is essential to the development of effective weather modification techniques. Such understanding would dictate, for example, the most effective method, location and time in the life cycle of the cloud for introduction of the seeding agent to produce the desired effect. Knowledge of the effects of small changes in the nuclei characteristics will permit a clearer evaluation of the inadvertent weather modification due to the release of pollution products into the atmosphere. Present theoretical models of cloud dynamics have included the microphysical processes in only the most rudimentary forms. This does not provide sufficient detail to adequately delineate the effect of the introduction of seeding agents. A knowledge of the roles of nucleation, activation and growth in the formation and development of clouds will permit

a more realistic incorporation of these vital parameters into the theoretical studies. To make such studies really valuable they must be integrated into a program designed to evaluate the interaction between the microphysics and the dynamics. This, in turn, should allow meteorologists to gain additional insight necessary for the improved acquisition and analysis of microphysical data in field experiments.

#### L.5 TERRESTRIAL LABORATORY LIMITATIONS

The use of simultaneously cooled walls in an expansion chamber, greatly extends the useful sensitive time of the chamber; however, conditions suitable for the study of cloud formation and development are still of limited duration. The cooled wall in conjunction with zero-g effectively eliminates two of the major limiting factors encountered in the use of fast expansion chambers, namely, heat conduction from the walls which is accompanied by loss of adiabaticity and the rapid development of gravity-induced convection. The latter factor, while it can be greatly suppressed, cannot be totally eliminated in terrestrial chambers. From a practical standpoint, it is almost impossible to construct a usable expansion chamber in which the walls have a perfectly uniform temperature, particularly in the vicinity of the inlet ports and observation windows required for access to the sensitive volume. The resulting temperature variations serve as a source of gravity-induced convection currents which can gradually build up during the course of the experiment until they cause disturbances which exceed acceptable levels. When the temperature in the chamber decreases with height, simulating a temperature inversion, convection can be partially suppressed but for most studies of adiabatic cloud expansions this condition is undesirable. In zero-g much less stringent requirements are placed on the thermal uniformity of the walls since no convection will be induced.

The second limitation to the duration of an experiment in a terrestrial expansion chamber is sedimentation of both large precipitation particles and cloud drops. This has not been a major problem with fast expansion chambers in the past due to the extremely short sensitive times. An entire experiment must be concluded in a period on the order of a few seconds. With the extended sensitive times possible in cooled wall chambers, fallout becomes a very real

1

limiting factor due to the long duration of experiments. Retention times for drops of  $10\mu$  diameter, which have a fall velocity of approximately 1 cm/sec, are relatively short for chambers of a few feet in height. It should be stressed that the fall velocity increases as the square of the droplet diameter. The strong dependence of fall velocity on drop size further affects the developing cloud by preferentially removing the large drops causing an alteration of the droplet size distribution. While sedimentation is essential to such studies as collision coalescence, it makes studies of processes such as Ostwald ripening (growth after the expansion has stopped) impossible. To this day, no convincing evidence for Ostwald ripening under cloud conditions exists.

To overcome the limitations imposed by droplet fallout, the growth times must be shortened, but this may require the use of supersaturations which are larger than those occurring in nature. This, in turn, results in conditions which are not truly representative of the atmospheric processes. Some increase in experiment duration can be gained by extending the height of the chamber and limiting observations to the region near the bottom. Even though there are practical limits to this method, the earth bound cooled wall cloud simulation chamber possesses many unique qualities which should be exploited in studying in-cloud processes.

The restrictions imposed by gravity-induced convection and drop fallout limit the use of terrestrial expansion chambers to the initial stages of cloud formation and development if simulations are to be carried out using realistic time scales.

#### L.6 ZERO-GRAVITY OPPORTUNITIES

A zero-gravity environment is the one means by which the capability of the cooled wall expansion chamber can be greatly extended. Some of the restrictions on the simulation of adiabatic cloud expansions using cooled wall expansion chambers can be eliminated by operating such a chamber in zero-g. Both gravity induced convection and particle fallout disappear. While the thermal gradients, which act as sources of gravity-induced convection in terrestrial chambers, will also be present in a zero-gravity chamber, the absence of gravity takes away the driving force which induces convection.



The only air motion will be due to the expansion of the chamber or deliberately induced motions. The droplets, as they form, will be free-floating and subject only to motions caused by diffusion and Brownian forces, small inertial forces due to personnel movements in the spacecraft, or small air movements caused by expansion of the chamber.

The nucleation and growth processes occurring during the initial stages of cloud formation and development are not gravity-dependent and a zero-gravity environment can only aid in the studies by removing the previously discussed restrictions. This will permit the use of expansion rates and duration times which can be realistically compared to those occurring in the atmosphere.

Even in the later stages of cloud development the benefits of a zero-gravity environment are not lost because it is during this stage that Ostwald ripening may occur. Ostwald ripening, the growth of large drops at the expense of smaller ones, has been advanced as one mechanism by which the observed broadening of the drop size spectrum could occur. Terrestrial experiments have not been able to test this hypothesis due to removal of the large drops from the cloud by fallout before the effect can be observed. Therefore a zero-gravity environment represents the only means available to observe this possible important cloud or fog process.

Information concerning such gravity-dependent processes as collision coalescence, which occur in the later stages of cloud development, can be obtained by combining the results obtained from zero-gravity experiments with those obtained from similar experiments using larger terrestrial chambers.

#### L.7 QUANTIFICATION

While the use of a zero-gravity environment does not represent the only answer to the restrictions of gravity-induced convection and fallout on terrestrial chambers, it does represent a means of obtaining a ten year head start by circumventing some of the principal problems. Thermal gradients within the chamber would have to be reduced to levels far below those

currently achieved and at the same time the chamber height would have to be increased by at least an order of magnitude. This represents an unknown, large expenditure of time, money and manpower.

## L. 8 APPROACH

### L. 8.1 General

General comments on the operation of an expansion chamber system can be found under Approach 1 of Class 6 - Scavenging.

Studies of both the adiabatic cloud expansion simulation and terrestrial expansion chamber evaluation will utilize both condensation and ice nuclei in varying concentrations. The nuclei being studied will be introduced into the chamber at a known pressure, temperature, and relative humidity. The chamber will then be expanded and cooled at a rate which simulates the cloud updraft velocity to be studied. As the expansion proceeds, the concentration, position, and motion of the resulting drops and ice particles will be recorded by the time lapse camera (holography, if available) and the optical scattering measurements will provide information on droplet size and liquid water content as a function of time.

Due to the extension of the temperature range into the region of ice formation, efforts should be made to develop optical scattering techniques for ice particle size and solid water content. While these measurements would be extremely useful, their lack will not make it impossible to obtain much needed information from these experiments.

The different updraft velocities and cloud heights can be simulated by varying the expansion rate and duration. Other variations can include initial pressure, temperature, and relative humidity. Addition of the appropriate type and amount of pollution gases can be used to study the effects of these factors on cloud formation and development. The existence of the Ostwald ripening process can be tested by holding the chamber in the expanded position and observing the changes in drop size distribution with time.

The terrestrial expansion chamber evaluation involves performing an experiment in the terrestrial laboratory which is as nearly identical to the zero-g experiment as possible and then comparing the results of the two experiments to determine the effects of gravity on chamber operation. In many respects this will represent an extra benefit of the adiabatic cloud expansion simulation experiment since the terrestrial experiments will have to be conducted as part of the development and testing program.

### L. 8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	
Type	3
Pollutant	5
Pressure	
Temperature	3
Relative humidity	3
Charge	
Rate of cooling	3
Time	3
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

## L.9 PROCEDURE

All electrical components are turned on and checked to insure they are operational and the control and measuring circuits are checked and calibrated, if necessary. After the system has been checked all components except the computer can be placed on standby or turned off if power conservation is necessary.

The main noncondensable sample gas supply and the sump pump are turned on, the expansion chamber ports opened and the system purged to insure removal of particles left from previous experiments. While the system is being purged the control program for the intended experiment is loaded into the control computer and the time lapse camera is loaded. When the system has been purged the expansion chamber ports are closed and the thermal controls for the expansion chamber, humidifier and conditioning chamber turned on.

The flow of sample gas to the humidifier and aerosol generator are adjusted to the required flow rates. The aerosol generator and pollution gases, if used, are turned on and adjusted. The particle size, mass and concentration meters along with the hygrometer are turned on and the sample monitored as it leaves the conditioning chamber. Monitoring continues with adjustment of the humidifier temperature and aerosol generator until a usable sample results.

The expansion chamber ports are opened and the chamber purged with the sample. Particle and relative humidity monitoring is transferred to the expansion chamber outlet and the purge continued until the sample becomes stable with respect to time. The expansion chamber ports are then closed.

When the expansion chamber ports are closed the expansion mechanism, expansion chamber pressure control, laser, liquid water meter, drop size meter, video system and expansion chamber gas temperature are all turned on, and the chamber held at the initial pressure while it comes to thermal equilibrium. While the expansion chamber is coming to thermal equilibrium the aerosol generator, pollution gases, hygrometer, particle size meter,

particle mass meter, and thermal controls for the humidifier and conditioning chamber are turned off. The system, except for the expansion chamber, is then purged with clean gas to remove the remaining aerosol and pollution gases. The noncondensable gas supply and sump pump are then turned off.

When the expansion chamber has reached thermal equilibrium the time lapse camera is started and the preprogrammed expansion/cooling cycle initiated. During the expansion/cooling cycle the chamber is monitored with the video system and data from the pressure, temperature, liquid water and drop size sensors recorded. At the completion of the expansion/cooling cycle the time lapse camera is stopped. The pressure control, expansion mechanism, laser, liquid water meter, drop size meter, video and expansion chamber gas temperature are also turned off. The chamber is heated to  $+10^{\circ}\text{C}$  to melt any ice on the walls and then the thermal control is turned off.

The expansion chamber ports are opened, the noncondensable gas supply and sump pump turned on, and the chamber purged to clean it. While the chamber is being purged the time lapse camera is unloaded and the film stored. The computer data is examined to detect any major malfunctions and then transferred to permanent storage. The expansion chamber ports are reclosed, the gas supply and sump pump turned off. The rest of the system is then either shut down or preparations started for the next experiment.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on all electrical components	5
• Check operational readiness of components	5
• Check and calibrate control and measuring circuits	10
• Turn on noncondensable gas supply and sump pump	0.5
• Open expansion chamber ports	0.5
• Purge system	10
• Load control program into control computer	(5)
• Load time-lapse camera	(3)
• Close expansion camera ports	0.5
• Turn on expansion chamber thermal control	
• Turn on humidifier thermal control	0.5
• Turn on conditioning chamber thermal control	
• Adjust gas flow to humidifier	0.5
• Adjust gas flow to aerosol generator	0.5
• Turn on aerosol generator and pollution gases	1
• Turn on aerosol particle size spectrometer	0.5
• Turn on nuclei mass meter	0.5
• Turn on hygrometer	0.5
• Monitor sample relative humidity, nuclei size, nuclei concentration and nuclei mass	10
• Adjust humidifier temperature	(-)
• Adjust aerosol generator	(-)
• Open expansion chamber ports	0.5
• Transfer sample monitoring point for conditioning chamber outlet to expansion chamber outlet	0.5

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Monitor sample	8
● Close expansion chamber ports	0.5
● Turn on expansion mechanism, expansion chamber pressure control, laser, liquid water meter, drop size meter, video and expansion chamber gas temperature	1
● Let expansion chamber come to thermal equilibrium	10
● Turn off aerosol generator, humidifier and conditioning chamber thermal controls, particle size, mass and concentration meters, hygrometer and pollution gases	(2)
● Purge system	(4)
● Turn off noncondensable gas supply and sump pump	(1)
● Turn on time lapse camera	0.5
● Start expansion/cooling program	30
● Stop time lapse camera	0.5
● Turn off expansion control, expansion mechanism laser, liquid water meter, drop size meter, video and expansion chamber gas temperature	1
● Heat expansion chamber to +10°C and turn off expansion chamber thermal control	0.5
● Open expansion chamber ports	0.5
● Turn on noncondensable gas supply and sump pump	8
● Purge chamber	(3)
● Unload time lapse camera and store film	(4)
● Check computer data	(1)
● Transfer data to permanent storage	0.5
● Close expansion chamber ports	0.5
● Turn off noncondensable gas supply and sump pump	10
● Shut down system	



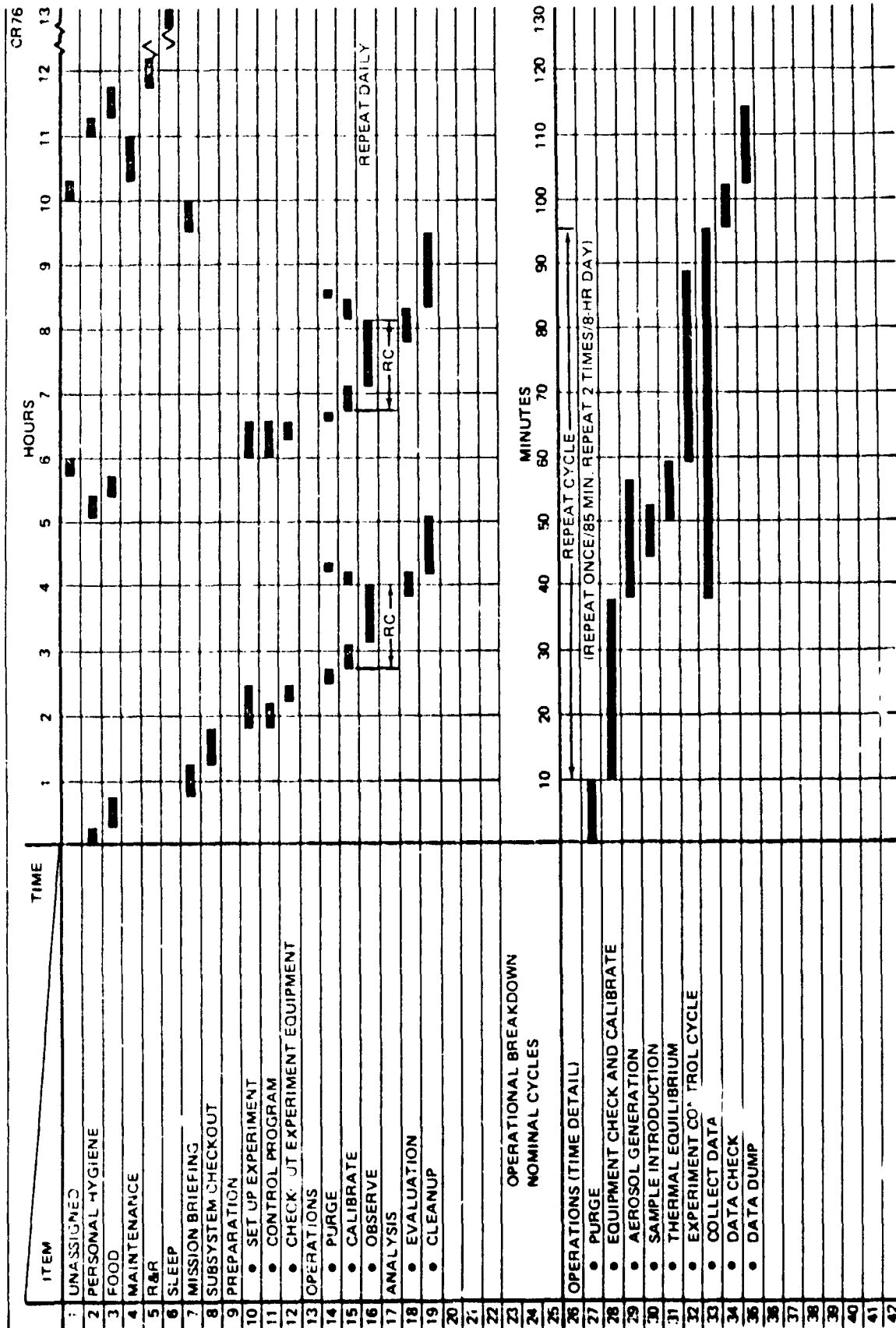


Figure L-1. Activity Timeline (One Day) Experiment Class 12

#### L.10 MISSION TIMELINE

At the present time, until a more detailed knowledge of the techniques that will be used in the final version, the activity timeline for the zero-g experimental procedure is the same as that developed for the terrestrial experiment (see Figure L-1).

It is possible that purge times and rates can be reduced but this can be determined only by testing of the actual equipment. The degree of automated control will affect the time required for some operations in both the zero-g and terrestrial experiments.

The primary change in the timeline for zero-g operation will be an extension of the duration of the expansion/contraction cycle to make full use of the zero-g environment.

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Appendix M  
CLASS 13  
ICE NUCLEI MEMORY

M.1 ICE NUCLEI MEMORY APPROACH 1

M.1.1 Introduction

The formation of ice phase in the atmosphere and particularly in supercooled clouds is of great importance in understanding the evolution of the clouds and in modifying them. In order to estimate the number of ice nucleations in the clouds, the concentration of ice nucleus particles is normally measured by introducing warm air samples into proper cold chambers to avoid ice crystals of other origins. However, it is known that some ice nuclei can retain "memory" on their surfaces under dry condition if ice nucleation has previously occurred on them (Fournier d'Albe, 1949; Mossop, 1956; Mason and Maybank, 1958; Serpolay, 1959) or the particles experienced a very low temperature (Higuchi and Futkuta, 1966). The formed memory allows the nuclei to form ice crystals at higher temperatures. Since it is the number of ice crystal formations, in terms of activation temperature and other variables, that is important in the cloud processes where ice phase is involved, the memory effect of ice nucleation needs to be clarified. The memory effect disappears if the sample air is warmed before testing as commonly done.

Concerning the memory effect of ice nucleation, the proposed mechanisms are controversial (see Fukuta, 1966; Roberts and Hallett, 1968; Edwards and Evans, 1971). Fukuta suggests a capillary mechanism for the memory effect occurring in a strongly cooled but dry sample, although he does not deny the possibility of the surface memory mechanism first pointed out by Mason (1950). Roberts and Hallett, and Edwards and Evans support only the surface memory mechanism or two-dimensional memory mechanism.

#### M.1.2 Objective

This study is therefore directed to clarify the controversial memory mechanism utilizing the unique zero-gravity condition in the space laboratory.

#### M.1.3 Scientific Justification

The study is important in order to clarify true mechanisms of memory retention which permit us to comprehend in overall memory effect of solid formation kinetics in general chemical substances. As the current nucleation theory takes the surface and bulk free energies into account for cluster formation in the fluctuating volume, it will be an applicability test of the theory under capillary conditions. Had the theory failed, it would warn us of the existence of a gap in our knowledge and suggest a necessary area of further study. The study will also provide some knowledge about the structure and behavior of water molecule clusters on the nucleus surfaces.

The scientific justification in terms of possible application is given below.

#### M.1.4 Applications

The obtained data will permit one to estimate ice nucleation in clouds and fogs more accurately. Particularly pertinent are survival of cirrus ice crystals and ice nuclei and their seeding of clouds below, and creation of the gap between ice crystals and ice nuclei observed in some clouds.

#### M.1.5 Terrestrial Laboratory Limitations

Two possible places for memory retention are capillary and surface, and the meaningful test of the phenomenon requires a condition clearly free of one of them. As the latter, i.e., surface, cannot be eliminated, the only choice is to produce and utilize a capillary-free condition. Aerosol particles formed by vapor condensation and before coagulation are a possible choice, but they tend to settle due to the gravitation in terrestrial laboratories. Supporting particles by a solid surface automatically introduce capillaries at their contact points.

#### M. 1. 6 Zero-Gravity Opportunities

The settling of aerosol particles as well as formed ice crystals presents a serious problem when the experiment cycling must be repeated more than once for the same aerosol sample. It may be expected that the number of ice nuclei with the surface memory will decrease in proportion to the smoke coagulation and the number with capillary memory will increase with respect to the extent of the coagulation, at least at the beginning. The particle settling acts toward reducing the number. If the settling is allowed, it induces an additional complication. The low gravity condition of the Zero-Gravity Cloud Experiment Laboratory is ideal and allows one to perform a clear-cut experiment without convection and particle fallout.

#### M. 1. 7 Approach

##### M. 1. 7. 1 General

Studies carried out so far on the memory effect in the terrestrial laboratories all employed substrates for ice nucleus particle support. As mentioned in (e) of Task 2, the capillary free condition for ice nucleation tests is the crucial requirement in order to carry out a decisive experiment for the mechanism. The Zero-Gravity Laboratory fortunately provides such an opportunity of particle suspension without any support.

A preliminary experiment showed that there was no sign of memory effect on 1, 5-dihydroxynaphthalene smoke (Fukuta, 1972) which is contrary to the claim by Edwards and Evans, although the activation temperature was rather high (-10 to -20°C).

Samples are mostly organic ice nuclei compounds, but some inorganic compounds such as lead and silver iodide can be used. Soil samples of various kinds should also be tested. The soil sample test allows checking if there is any memory effect in freely suspended particles in air but does not serve the purpose of distinguishing the possible mechanism of the memory effect.

The experimental procedure is as follows. Organic smoke particles prepared by a LaMer-Sinclair Generator or the Denver University smoke generator are introduced into the smoke chamber. The smoke particles in the chamber will

age by coagulation with each other and with the wall. The smoke concentration should be about  $10^5$ /cc so that the coagulation proceeds at a proper rate. At a given time interval, approximately an hour, a small amount of smoke sample is introduced into the cold expansion chamber. The expansion chamber is kept at a subfreezing temperature  $T_1$  with a supercooled fog created by injecting the steam from the steam source. The ice nucleation proceeds in the fog. After photographing and confirming the number concentration of nucleated ice particles by the light beam method (illuminate a small, known volume of the cold chamber and count the number of ice crystals with the naked eye), the chamber is quasi-isothermally and therefore relatively slowly expanded by maintaining the same temperature at the wall until the pressure reaches a predetermined value  $P''_1$  from the initial value of  $P_1$ . During this course, both the total pressure of the chamber at which the ice crystals have just sublimed,  $P'_1$ , and the temperature of air,  $T'_1$ , will be measured. This point is under ice saturation. Therefore, from  $P'_1$ ,  $T'_1$ ,  $P''_1$ , and  $T_1$  one can estimate the relative humidity of the air (RH) at  $P''_1$  position. This dry condition of the chamber will last for about 20 minutes. Then the chamber wall temperature will quickly be raised slightly above the threshold temperature of the ice nucleation,  $T_2$  (about  $2^\circ\text{C}$  warmer than the threshold), and the chamber air will slowly be compressed until the predetermined ice saturation point is reached at temperature  $T_2$ . A small amount of moisture will be introduced from the steam source and the number of ice crystals formed in the given volume of the light beam will be counted.

If any ice crystals are detected here, it is a sign that the memory exists. In order to confirm the memory effect, the air will be heated up to a temperature  $T_3$ , say to  $10^\circ\text{C}$ , by a rapid adiabatic compression coupled with the chamber wall warming. After holding the warm condition for one minute, the system will be quickly cooled back to the previous condition again quickly by adiabatic expansion coupled with the cooling. The number of ice crystals formed will be checked and compared with that found before this warming process.

After this, the chamber air will be replaced with clean, filtered air and be ready for the next run.

The same experiment will be repeated for a duration sufficient to determine whether the ratio between the number of ice nucleation by memory effect and that of the first nucleation increases with respect to time. Such an increase is a sign of a capillary memory effect.

The chamber temperature needs to be reduced to a level as low as  $-60^{\circ}\text{C}$ . The pressure will have to be lowered at least to  $1/2$  atmosphere. One experimental run will take several hours due to the required aerosol aging. For this study there are three main factors under which the experimental runs will have to be made (i.e.,  $T_1$ , RH at  $P''_1$ , and the sample). The suggested ranges of the variables are:

$T_1$ :  $-60$ , and  $(T_c - 3)^{\circ}\text{C}$

$T_c$ : the nucleation threshold temperature

RH: 0 and 100% with respect to water

Sample: 1, 5-Dihydroxynaphthalene and Plexiglas Powder (non-ice nucleating) for organics,  
AgI for inorganics  
and one soil sample

To save time, two relative humidity levels may be taken alternately for the runs in the same day.

#### M.1.7.2 Instrumentation

In order to clarify the mechanism, it is necessary to create a capillary-free condition and test the memory effect in it. If the nucleus compound is supported by a surface, capillaries form at points on contact. Therefore, the compound must be suspended in air in order to avoid capillary formation. The compound should not carry any capillaries in itself, at least at the beginning, and the smoke particles formed by slow condensation should satisfy this requirement if they are kept apart to prevent the coagulation. Once the capillary-free particles of ice nuclei are formed, it would be easy to conduct a suitable experiment making use of the expansion chamber with additional vapor supply for mixing and a simple removable stirrer.

A simplified LaMer-Sinclair Monodispersed Aerosol Generator can be used for smoke particle generation. This generator consists of a nuclei source (normally a heated nichrome wire) evaporator of nuclei compound with



dilution gas inlet, mixing mechanism of the nuclei and vapor, and a cooling tube for gentle nucleation and growth. This generator uses a molten chemical to increase the vapor pressure. It may be necessary to use woven fiber glass to restrain the liquid in order to prevent the liquid from floating.

Each compound uses its own set of an evaporation chamber and a cooling tube. The set has to be made interchangeable so that it easily connects to the all purpose nuclei source and to tubings leading to the smoke chamber and the smoke filter system.

For preparation of a Plexiglas sample, a heating tube is necessary instead of a cooling tube. The Plexiglas powder will be sent out of a squeeze bottle and when the powder particles pass through the heating tube, they melt, creating spherical particles free from capillaries.

The smoke chamber should be about  $1\text{ m}^3$  in size with inlets for clean air and sample smoke, a port for sampling and an outlet to the smoke filter. After filtration of used smoke, the air will be recycled. The cleaning can be done by sucking the smoke air through a tightly packed layer of cotton and an activated charcoal layer. Cleaning of chamber walls will be made by a vacuum cleaner.

The temperature controlled expansion chamber requires a vapor supply. The vapor supply is a flask in which water in a semipermeable tube is kept warm by an electric heater.

Other equipment necessary for the study besides the main expansion chamber are a mercury lamp for chamber illumination, and a camera for photographing ice crystals which have formed.

#### M. 1. 7. 3 Measurement and Data Requirements

Photographs will provide data storage of the quantities of ice crystals versus various chamber condition cycles. Commentary will be recorded at appropriate points during the experiment along with digital recording of time, temperature, and pressure.

#### M.1.7.4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	1
Size-droplet	
Type	4
Pollutant	
Pressure	8
Temperature	8
Relative humidity	6
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

### M.1.8 Procedure

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with thought to translating this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient (see Figure M-1).

<u>Activities</u>	<u>Minutes</u>
• Generate nuclei within smoke chamber	20
• Purge expansion chamber	5
• Cool expansion chamber to subfreezing temperature (Figure M-2)	20
• Establish pressure $P_1$	
• Generate supercooled fog using vapor supply	5
• Inject nuclei sample	5
• Photograph nucleated droplets and following events	
• Quasi-isothermal expansion to pressure $P''_1$	20
• Record temperature and expansion	
• Raise chamber wall temperature to $T_2$	
• Compress chamber to $P_2$	10
• Introduce moisture and photograph resulting ice crystals	3
• Raise temperature to $T_3 (+10^\circ\text{C})$ by rapid compression	1
• Expand to pressure $P_2$ , temperature $T_2$	
• Photograph resulting ice crystals	
• Recycle with another aged nuclei sample	
• Repeat for other nuclei types	

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● Generate nuclei within conditioning smoke chamber Smoke particle concentration check by an ultramicroscope Smoke concentration adjustment by dilution with clean air Use 1 m<sup>3</sup> smoke chamber</li> </ul>	20
<ul style="list-style-type: none"> <li>● Purge expansion chamber</li> </ul>	5
<ul style="list-style-type: none"> <li>● Cool expansion chamber to subfreezing temperature T<sub>1</sub> (Figure M-2).</li> </ul>	20
<ul style="list-style-type: none"> <li>● Establish pressure P<sub>1</sub></li> </ul>	
<ul style="list-style-type: none"> <li>● Generate supercooled fog using vapor supply</li> </ul>	5
<ul style="list-style-type: none"> <li>● Inject nuclei sample</li> </ul>	5
<ul style="list-style-type: none"> <li>● Photograph ice crystals and following events (2 photos)</li> </ul>	
<ul style="list-style-type: none"> <li>● Quasi-isothermal expansion to pressure P<sub>1</sub>"</li> </ul>	20
<ul style="list-style-type: none"> <li>● Record temperature and expansion</li> </ul>	
<ul style="list-style-type: none"> <li>● Raise chamber wall temperature to T<sub>2</sub></li> </ul>	10
<ul style="list-style-type: none"> <li>● Compress chamber to P<sub>2</sub></li> </ul>	
<ul style="list-style-type: none"> <li>● Introduce moisture from vapor supply and photograph resulting ice crystals (2 photos)</li> </ul>	3
<ul style="list-style-type: none"> <li>● Raise temperature to T<sub>3</sub> (+10°C) by rapid compression to P<sub>3</sub></li> </ul>	1
<ul style="list-style-type: none"> <li>● Expand to pressure P<sub>2</sub>, temperature T<sub>2</sub></li> </ul>	2
<ul style="list-style-type: none"> <li>● Photograph resulting ice crystals (2 photos)</li> </ul>	
<ul style="list-style-type: none"> <li>● Recycle with another aged nuclei sample</li> </ul>	
<ul style="list-style-type: none"> <li>● Repeat for other nuclei types</li> </ul>	

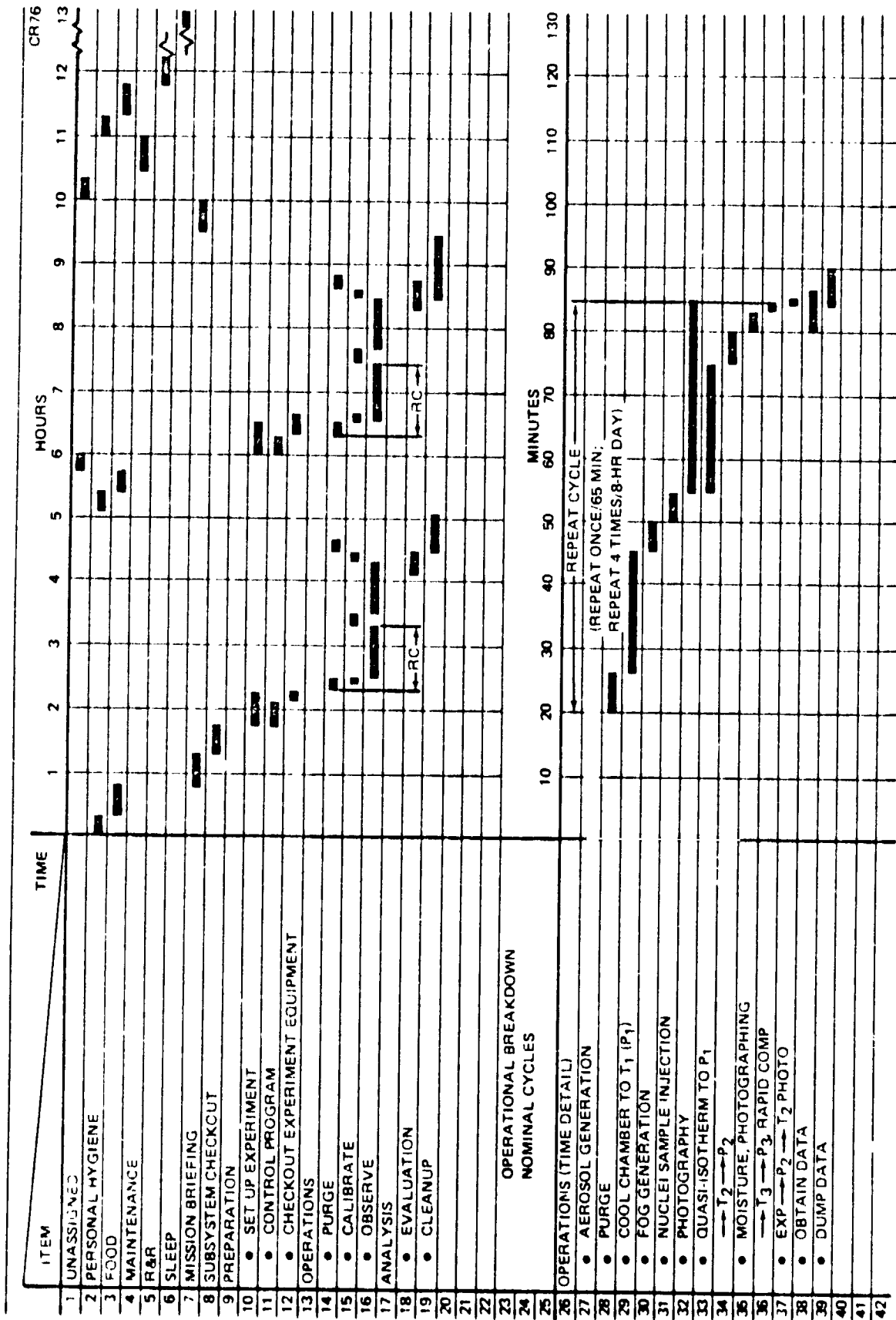


Figure M-1. Activity Timeline (One Day) Experiment Class 13

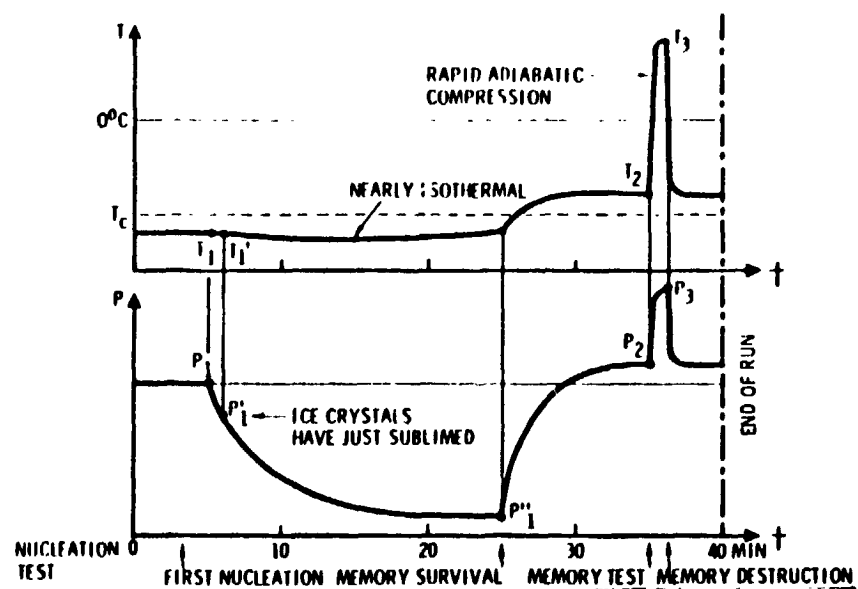


Figure M-2. Process of Adiabatic Expansion Chamber for Memory Study

#### M.1.9 References

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## **M.2 ICE NUCLEI MEMORY APPROACH 2**

### **M.2.1 Introduction**

There has been significant concern expressed by the scientific community<sup>1</sup> because ice nuclei counts at cloud base fail to systematically correlate with ice crystal counts in the upper regions of the cloud. As pointed out under Experiment Class No. 2, Ice Nucleation, this discrepancy might arise from several different mechanisms which are poorly understood: (a) turbulent mixing introducing nuclei from different levels, (b) ice memory effects whereby ice crystals ejected and evaporated at cloud top become a source of preactivated nuclei which may be re-entrained into the cloud by providing a feedback mechanism which is highly dependent upon the state of glaciation of the cloud, (c) unknown factors involved in the method of measuring the ice nucleus concentration and evaluating their effectiveness in nucleating ice under the specific conditions prevailing in a particular cloud (see Experiment Class No. 2 for a more complete discussion of this question).

Gagin<sup>2</sup> suggests that there is no visible evidence for lateral mixing at cloud tower edges. Warner<sup>3</sup>, however, notes that, even in warm, non-precipitating maritime cumuli, the bimodal character of the drop size distribution is readily explained in terms of the mixing of parcels, presumably from the top of bottom of the cloud. He, too, found no convincing evidence for mixing from the peripheral edges of the cloud. Simpson<sup>4</sup>, on the other hand, agrees that turbulent mixing must play an important role in determining the state of cloud towers. The mechanisms involved in these feedback processes are highly dependent upon the nature of the ice memory effect which might exist.

### **M.2.2 Objective**

The objective of this experiment is to determine the effect of an ice nucleus' history on its ability to initiate (nucleate) the ice phase.

### **M.2.3 Scientific Justification**

The memory effect in the case of ice nuclei has not been studied nearly as extensively as the memory effect for Aitken sized particles. Many aspects have not been studied quantitatively. Although it is readily conceded that ice



crystals which once contained ice nuclei leave behind a residual ice nucleus upon evaporation which may be regarded as preactivated under certain conditions, little information is available about a possible memory effect associated with the evaporation of ice crystal fragments which do not possess a particulate nucleus. Cwilong<sup>4, 5</sup> did a series of experiments in which ice crystals were nucleated homogeneously in an expansion chamber. He later evaporated these crystals and found that the system nucleated ice much more easily thereafter. This would tend to indicate that a memory effect exists for the evaporation of ice crystals regardless of whether or not the original ice crystal contained a particulate nucleus. The most pertinent question remains - at what temperature does such an ice memory effect disappear?

#### M. 2. 4 Terrestrial Laboratory Limitations

A large portion of the work done on ice nuclei has made use of capturing the aerosol particles on membranes and filters and then processing or developing these to determine the activity of the particles as ice nuclei. Because the mechanics of the aerosol is stopped, it is difficult to mimic such features as contact nucleation, although Langer has bombarded the aerosol-laden filters which supercooled water droplets in an attempt to determine the number of nuclei which are capable of responding to this mechanism. Another feature of membranes is that they produce a host of extraneous effects as a result of the surface which connects the particles. At high relative humidities, thick films of adsorbed water can transport surface active agents relatively easily, a fact which accounted for the collection of contaminated water in capillaries under the guise of anomalous water. The introduction of surface transport makes the kinetics of nucleation on surfaces different than nucleation in suspended systems.

In a cloud chamber system, the time required to perform a complete cycle consisting of nucleating ice crystals, melting or evaporating them, and then attempting to evaluate the ice nucleating capability of the residual aerosol, precludes the feasibility of performing such experiments in a terrestrial laboratory facilities of moderate size. If chambers of extremely large size are used, additional problems are introduced in maintaining homogeneity.

#### M. 2. 5 Zero-Gravity Opportunities

The advent of the zero-g opportunity now makes it possible to carry out experiments which require relatively long time intervals on suspended particulate systems. It is a convenient way of circumventing problems associated with suspending particles on films and wires. It also alleviates problems associated with going to extremely large cloud chambers. Experiments can be carried out in a chamber of convenient dimensions and the particles will remain relatively stationary throughout the experiments. Observational systems need not have the capability of sampling particles in many widely separated areas in the chamber in order to obtain a suitable spatial average throughout a large chamber.

#### M. 2. 6 Quantification

Ice nuclei will be introduced into the chamber at a specific relative humidity. If some of the nuclei are effective as condensation nuclei, both a water cloud and an ice crystal cloud will be formed upon simulation of the updraft. The cloud drops will provide water saturation at whatever degree of supercooling is established by the cloud chamber program. The chamber may be adiabatically recompressed and the ice crystals will melt. If the chamber is re-expanded, it can be determined whether the same concentration of ice crystals forms once again as a result of a memory effect associated with the melting of the ice crystals.

#### M. 2. 7 Approach

##### M. 2. 7. 1 General

General comments on the operation of an expansion chamber system can be found under approach 1 of Class 6 - Scavenging.

In studying the memory effects of ice nuclei, the nuclei should be subjected to three different sequences of environments after the initial ice nucleation has taken place. In the first sequence, the nuclei are dried while being maintained at subfreezing temperatures and then tested by nucleating the ice phase. In the second, the nuclei are also dried, but the temperature if permitted to go above freezing before renucleating the ice phase. The third

sequence does not dry the nuclei but only raises their temperature above freezing before nucleating the ice phase.

The initial ice formation is achieved by placing a mixture of condensation and ice nuclei into the expansion chamber at a known pressure, temperature, and relative humidity. The chamber is then cooled by an adiabatic expansion. The initial conditions are chosen such that a cloud will form on the condensation nuclei at temperatures slightly above those at which the ice nuclei are active. As the cloud of water drops is supercooled, the ice nuclei start to cause ice nucleation through all three mechanisms: contact freezing, internal freezing, and vapor deposition. After the chamber has been cooled to a predetermined temperature, the expansion is stopped and the chamber held at the final temperature to give the ice nuclei time to activate and the ice particles to grow.

When considering methods available for decreasing the relative humidity sufficiently to evaporate the ice particles and leave only the nuclei, the first method that comes to mind is use of an adiabatic compression. The problem with this technique is that not only is the relative humidity decreased but the temperature is simultaneously increased and for the vast majority of cases of interest the temperature is raised considerably above 0°C which should destroy any memory effect for ice nuclei.

A second technique considered is the use of an isothermal expansion to decrease the vapor pressure below the ice saturation pressure at the temperature of interest. This method appears promising until one considers the amount of water vapor present after the drops and ice particles have been evaporated and the volumetric expansion ratios required to lower the vapor pressure to ice saturation. For example, to change a cloud of one hundred  $10 \mu$  drops per  $\text{cm}^3$  to vapor at the saturation pressure of ice at -40°C requires an expansion ratio in excess of 4. And this is only if the drops represent the total initial water content of the chamber; any other situation increases the required expansion ratios even more.

The only feasible method is to remove the sample of drops and ice particles from the expansion chamber, pass them through an external drying chamber, and return the dried sample to the expansion chamber using a closed loop. To prevent destruction of the memory effect, the entire external sample loop must be maintained at the same temperature as the interior of the expansion chamber.

Once the nuclei have been dried, they must be subjected to a second supersaturated environment to measure the changes in their nucleation characteristics indicating the degree of memory exhibited by the nuclei. Unfortunately, the drying process will have removed all water vapor from the sample and when the chamber is warmed and compressed to the initial temperature and pressure of the test expansion the chamber will be highly subsaturated. Therefore, the water vapor must be replaced by circulating the sample through the external loop, but this time the desiccating chamber is replaced by a humidifier. After the sample has been rehumidified, the expansion chamber is resealed and a second adiabatic expansion used to reactivate the ice nuclei. As previously stated, the changes in the ice nucleating characteristics of the nuclei between the first and second expansions can be related to memory effects.

The initial temperature of the second expansion should be varied over a range which extends from well below freezing ( $-15^{\circ}\text{C}$ ) to above freezing ( $+10^{\circ}\text{C}$ )

The final test would be to use an adiabatic compression to raise the temperature of the nucleated ice crystals above  $0^{\circ}\text{C}$  sufficiently to insure melting and then observe the refreezing process.

#### **M. 2. 7. 2 Experiment Parameters**

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	
Type	2
Pollutant	
Pressure	
Temperature	9
Relative humidity	3
Charge	
Rate of cooling	2
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

### M.2.8 Procedure

Initially, all the electrical components are turned on and checked for operational readiness, then the control and measuring circuits are checked and calibrated if necessary. The noncondensable gas supply and sump pump are turned on, the expansion chamber ports opened and the system cleaned by purging. While the system is being purged, the time lapse camera is loaded and the control program loaded into the control computer.

When the cleaning purge has been completed the expansion chamber ports and external sample circuit are closed. The thermal controls for the humidifier, conditioning chamber, expansion chamber and external sample circuit are turned on. The gas flow to the humidifier and aerosol generators are adjusted and the aerosol generators turned on. The hygrometer, particle size and mass meters are turned on and the sample monitored for relative humidity, particle size, mass and concentration as it leaves the conditioning chamber. The humidifier and aerosol generators are adjusted until the sample has the required characteristics and has reached equilibrium.

The expansion chamber ports are opened and the sample introduced into the chamber. Sample monitoring is transferred to the expansion chamber exhaust port and sample introduction continued until equilibrium is reached. The expansion chamber ports are then closed. The expansion mechanism, pressure control, gas temperature, video, laser, liquid and solid water meters, and drop and ice particle size meters are turned on. The expansion chamber is then allowed to come to thermal equilibrium.

While the expansion chamber is coming to thermal equilibrium the humidifier and conditioning chamber thermal controls, aerosol generators, hygrometer, and particle size and mass meters are turned off. The system, except for the expansion chamber and external sample circuit are then purged to remove the remaining aerosol. The noncondensable gas supply and sump pump are then turned off.

When the expansion chamber has reached thermal equilibrium the time lapse camera is turned on and the initial adiabatic expansion started. The chamber is observed as it is cooled and expanded to a preset final temperature and then held while ice formation occurs. The chamber is then opened to the external sample circuit, which has also been cooled to the same temperature as the expansion chamber, and the diaphragm pump is turned on to circulate the sample from the expansion chamber through the drier and back to the expansion chamber. After the sample has been dried, the diaphragm pump is turned off while the expansion chamber and external sample circuit are warmed and compressed to the test temperature by a quasiadiabatic compression. The diaphragm pump is restarted and the sample circulated through the humidifier to restore the water vapor content. When the sample has been rehumidified the diaphragm pump is turned off and the expansion chamber ports closed. The first search expansion is now used to test and evaluate the memory effects.

At the completion of the first search expansion, the chamber is warmed to a temperature far enough above freezing so that any ice memory effects should be destroyed. A second search expansion is then used to test for destruction of the memory effect.

At the completion of the second search expansion, the time lapse camera, expansion mechanism, pressure control, gas temperature, laser, video, liquid and solid water meters, and drop and ice particle size meters are turned off. The chamber and external sample circuit are warmed to  $+10^{\circ}\text{C}$  and the thermal controls turned off. The expansion chamber ports and external sample circuit are opened, the noncondensable gas supply and sump pump are turned on and the system purged.

While the system is being purged the time lapse camera is unloaded and the film stored for processing. The computer data is scanned and transferred to permanent storage. The expansion chamber ports and external sample circuit are then closed, and the noncondensable gas supply and sump pump turned off.

The rest of the system is then either shut down and cleaned up or prepared for the next experiment.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Turn on all electrical components	5
● Check operational readiness of components	5
● Check and calibrate control and measuring circuits	10
● Turn on noncondensable gas supply and sump pump	0.5
● Open expansion chamber ports and external sample circuit	0.5
● Purge system	10
● Load control program into control computer	(5)
● Load time-lapse camera	(3)
● Close expansion chamber ports and external same circuit	0.5
● Turn on thermal controls: conditioning chamber humidifier expansion chamber, external sample circuit	0.5
● Adjust gas flow to humidifier	0.5
● Adjust gas flow to aerosol generators	0.5
● Turn on aerosol generators	1
● Turn on particle size spectrometer, hygrometer, particle mass meter	1
● Sample relative humidity, nuclei size, mass, concentration	10
● Adjust humidifier and aerosol generators as required	(-)
● Open expansion chamber ports	0.5
● Transfer monitoring to output of expansion chamber	0.5
● Monitor sample relative humidity, nuclei size mass, concentration	8
● Close expansion chamber ports	0.5
● Turn on expansion mechanism, pressure control, gas temperature video, laser, liquid water meter, drop size meter, ice particle size meter, solid water meter	1



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Let expansion chamber come to thermal equilibrium	10
● Turn off aerosol generators thermal controls for humidifier and conditioning chamber, particle size, mass and concentration meters, hygrometer	(2)
● Purge system (except expansion chamber)	(4)
● Turn off noncondensable gas supply and sump pump	(1)
● Turn on time-lapse camera	0.5
● Start cooling/expansion program	20
● Observe chamber on video monitor	-
● Stop cooling/expansion and hold	0.5
● Open expansion to external sample drier circuit	0.5
● Start diaphragm pump	0.5
● Circulate sample through drier	20
● Stop diaphragm pump	0.5
● Change flow from drier to humidifier	0.5
● Warm and compress expansion chamber and external sample circuit to test temperature ( $<0^{\circ}\text{C}$ )	20
● Start diaphragm pump	0.5
● Circulate sample through humidifier	20
● Stop diaphragm pump	0.5
● Close expansion chamber	0.5
● Start first search expansion/cooling	20
● Observe chamber on video monitor	(-)
● Warm/compress expansion chamber to $+10^{\circ}\text{C}$	20
● Start second search expansion/cooling	20

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Stop time-lapse camera	0.5
● Turn off expansion mechanism, pressure control, gas temperature video, laser, liquid water meter, drop size meter, ice particle size meter, solid water meter	1
● Heat chamber to +10°C. Turn off expansion chamber thermal control	5
● Open expansion chamber ports and external sample circuit	0.5
● Turn on sump pump and noncondensable gas supply	0.5
● Purge chamber and external sample circuit	8
● Unload time-lapse camera and store film	(3)
● Check computer data	(4)
● Transfer data to permanent storage	(1)
● Close expansion chamber ports	0.5
● Turn off noncondensable gas supply and sump pump	0.5
● Shut down system or start next experiment	10

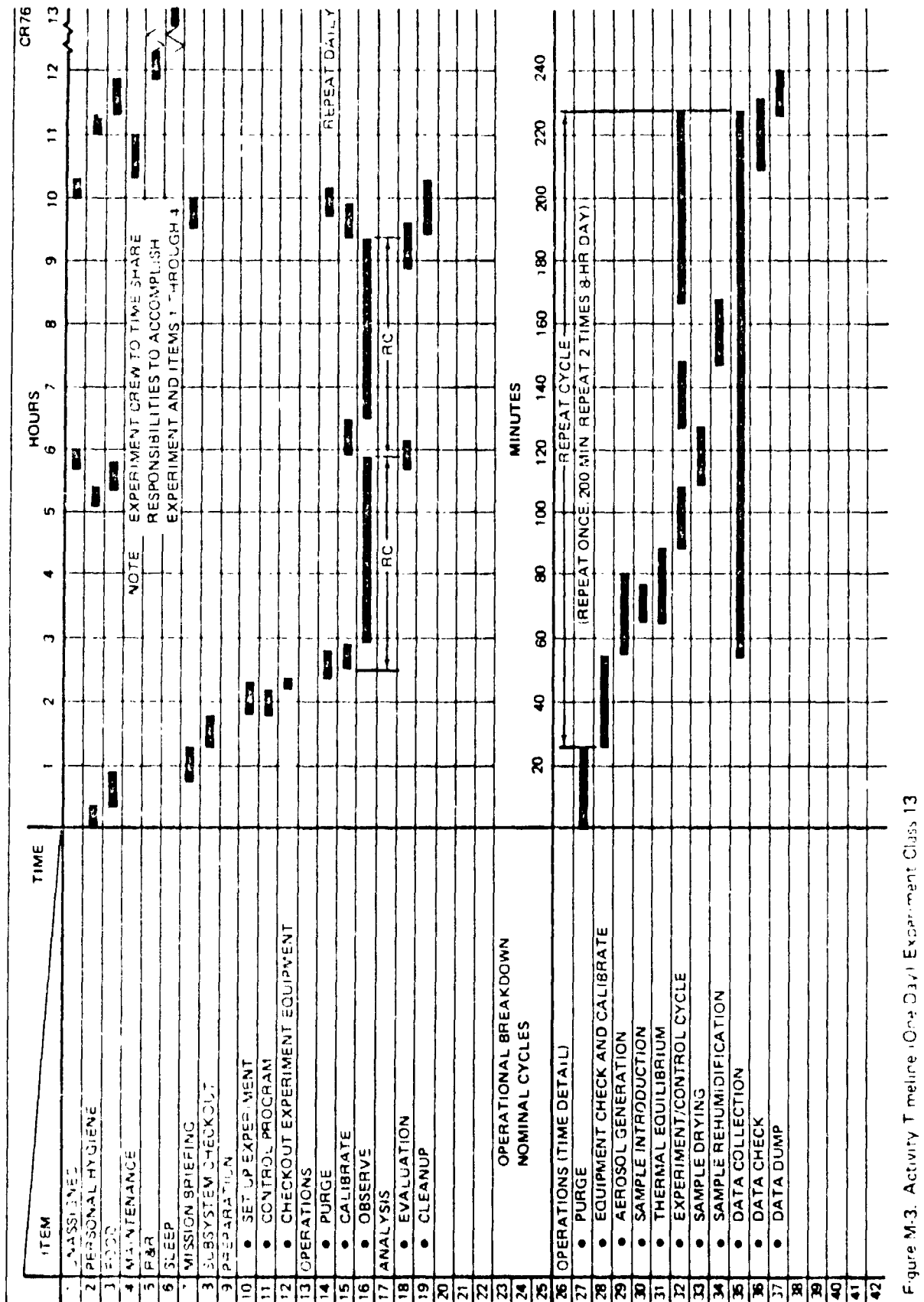


Figure M-3. Activity Timeline (One Day) Experiment Class 13

#### M. 2. 9 Mission Time Line

At the present time until a more detailed knowledge of the techniques that will be used in the final version the activity time line for the zero-g experimental procedure is the same as that developed for the terrestrial experiment (see Figure M-3).

It is possible that purge times and rates can be reduced but this can be determined only by testing of the actual equipment. The degree of automated control will effect the time required for some operations in both the zero-g and terrestrial experiments.

The primary change in the time line for zero-g operation will be an extension of the duration of the expansion/contraction cycle to make full use of the zero-g environment.

#### M. 2. 10 References

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2. A. Gagin, private communication (1973).
3. J. Warner, "The Microstructure of Cumulus Cloud. Part I. General Features of the Droplet Spectrum", Journal Atmospheric Sciences, 26, 1049-1059 (1969); "The Microstructure of Cumulus Cloud. Part II. The Effect on Drop Size Distribution of the Cloud Nucleus Spectrum and Updraft Velocity", Journal Atmospheric Sciences, 26, 1272-1282 (1969).
4. J. Simpson, private communication (1973).
5. B. M. Cwilong, "Sublimation in a Wilson Chamber", Nature, 155, 361 (1945); "Sublimation in a Wilson Chamber", Proc. Roy. Soc., A190, 137 (1947).

Appendix N  
CLASS 14  
TERRESTRIAL EXPANSION CHAMBER EVALUATION

N.1 INTRODUCTION

Most of the details for this class have been incorporated into the Class 12 description. The following are a few general comments from other sources.

The expansion chamber is a very important instrument that is used for the studies of ice and condensation nuclei properties which are present in the atmosphere. This chamber provides the necessary supersaturation for nucleation by adiabatic expansion cooling. Working above freezing provides information concerning the condensation nuclei which participate in the precipitation processes, while below freezing temperatures are used for ice nuclei studies. The characteristics of natural and artificial nuclei under representative atmospheric conditions must be known before cloud seeding can be used to redistribute rain and snow, diminish the damage due to hail and lightning, and moderate the effects of hurricanes. The effects of pollutants on atmospheric processes and man's health are also important and can be studied in the expansion chamber.

Most present expansion chambers are limited by convection to a few tens of milliseconds, whereas atmospheric-important processes range from a few tenths of a second to minutes in duration. This convection is a result of non-uniform cooling of the air near the chamber walls. Gravity then causes the heavier air to move downward, resulting in convection. Past attempts to cool the walls of the chamber have been made with little success. Chambers now under development promise to solve part of this convection problem as well as thermal diffusion problems by cooling the walls of the chambers at the same rate that the air is being cooled. If these chambers are successful, the observation times can be extended to a few seconds with this new limit being imposed by gravity-induced fallout. While this extension will provide much needed data, even longer times are needed.

## N.2 OBJECTIVE

The objective of this experiment is to measure condensation and ice nuclei activation efficiencies under operating conditions similar to those utilized in terrestrial laboratories, but within gravity-induced convection.

## N.3 APPLICATIONS

The expansion chamber has been used to study nucleation properties of condensation and ice nuclei. Corrections for the nuclei counts under gravity-induced convection and fallout would permit more accurate studies of atmospheric nuclei and how they participate in atmospheric precipitation processes. This knowledge would be used in weather modification efforts involving rain, snow, and fog.

## N.4 ZERO-GRAVITY OPPORTUNITIES

The low-gravity conditions of a space laboratory would reduce the convection and fallout limitations of an expansion chamber by an amount related to the reduction of the acceleration level. Experiments in these conditions would provide unambiguous numbers relative to specific expansion rates and initial and final conditions. These numbers can then be compared with terrestrially obtained data to determine errors due to convection and fallout. Using such a procedure of comparison for low-g and 1-g chamber results, correction factors can be obtained that would permit the expansion chamber to be operated at lower expansion rates which are more representative of atmospheric conditions (i. e., the useful range of an expansion chamber operating in a terrestrial environment can be extended).

## N.5 APPROACH

### N.5.1 General

Two (or pairs of) identical expansion chambers will be used, one operating in a terrestrial laboratory and the other operating in a low-gravity environment. A series of nucleation experiments would be performed in both chambers using standardized nuclei sources and following presently accepted operating procedures. Recommendations for terrestrial chamber modifications and operating procedures may result from the comparisons of these

chambers. Next the chambers will utilize slower expansions than are normally acceptable in a terrestrial laboratory. The results will be used to see if consistent correction factors can be applied to the terrestrial laboratory chambers so that some future experiments could be performed on the ground with repeatable results, rather than being conducted in space. These chambers will incorporate the latest expansion and cooled-wall techniques to obtain the maximum operating times.

Initial expansion chamber and supporting subsystems will be similar to those presently used in terrestrial laboratories. This similarity is needed to satisfy the goal of evaluating the numbers obtained in a terrestrial laboratory. Future designs for both terrestrial and low-gravity chambers will incorporate changes in geometry and procedure as improvements are specified. Present small chambers are usually cylindrical in form, about 30 cm in diameter and 45 cm in height. The initial pressure, temperature and relative humidity, and final pressures must be measured to accuracies of 0.05 percent or better. These requirements are pushing the state of the art, especially in the area of relative humidity or total water content measurements. Optical techniques utilizing light scattering and absorption techniques are being developed to detect the water content within the chamber. Optical scattering techniques are also being refined for the detection of submicrometer diameter particles within the chamber, thus monitoring their growth with time. Raman spectroscopy may permit quantitative monitoring of the gas composition for those experiments involving "pollution" gases.

#### N. 5.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	5
Size-droplet	
Type	5
Pollutant	
Pressure	
Temperature	

<u>Parameters</u>	<u>Variations</u>
Relative humidity	
Charge	2
Rate of cooling	4
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

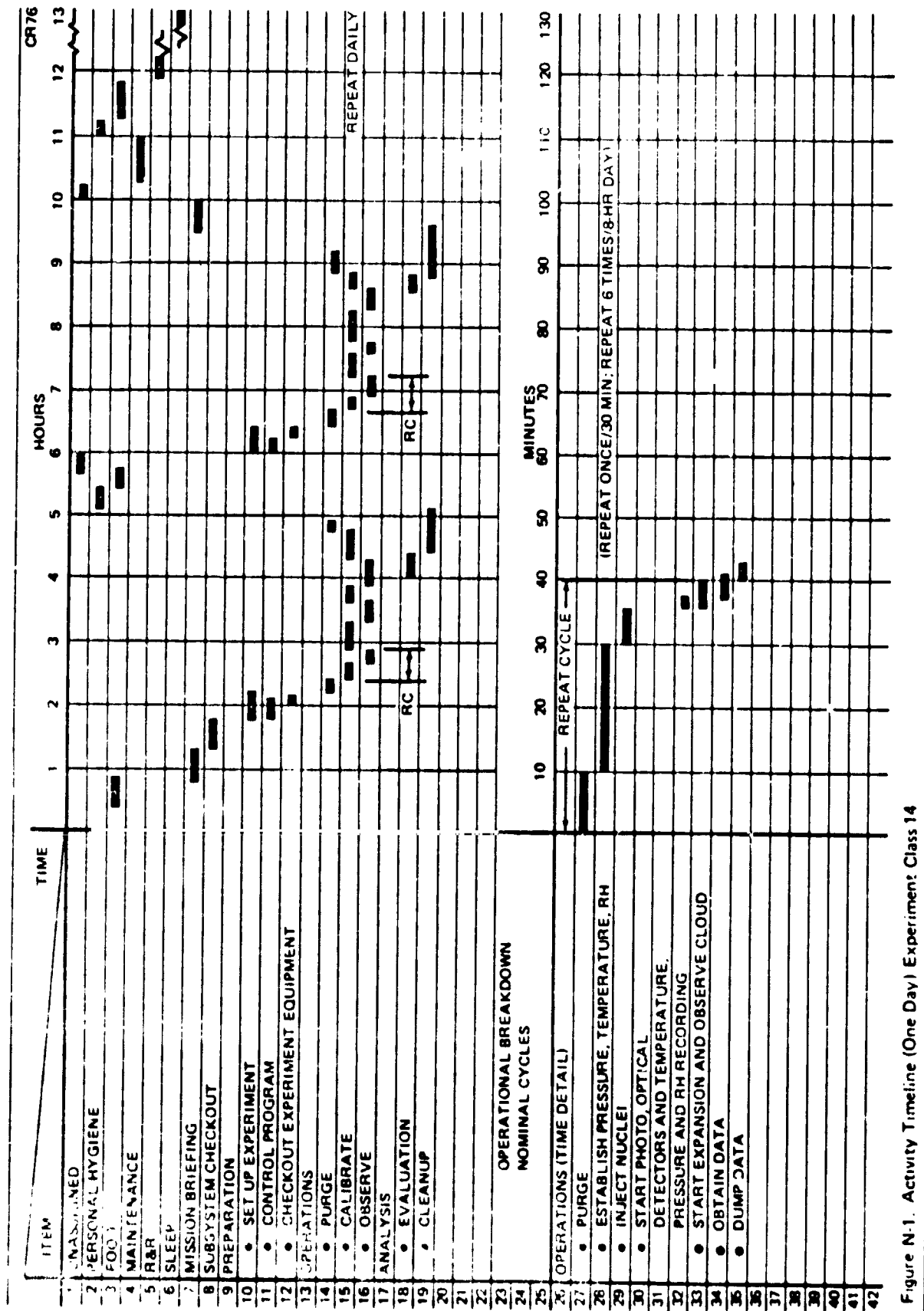
#### N.6 PROCEDURE

General activity details are given below followed by a representative activity timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure N-1).



# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Purge chamber	10
• Establish initial pressure, temperature and relative humidity	15
• Inject nuclei	5
• Start camera, optical detectors and T, P, RH recording	2
• Start expansion	
• Expand and observe formed cloud	30
• Compress and recycle and required	30
• Recycle with other expansion rates and final values	
• Recycle with other initial T, P, RH values	
• Recycle with other nuclei concentrations and types	



Appendix O  
CLASS 15  
CONDENSATION NUCLEI MEMORY

O.1 CONDENSATION NUCLEI MEMORY APPROACH 1

O.1.1 Introduction

It is known that cloud condensation nuclei (CCN) behave differently after the cycle of cloud droplet formation followed by evaporation. This effect may partially be explained by the surface effect (Smith, et al., 1968) which merely slows the growth or evaporation rate of cloud droplets depending on the size of nuclei and the thermal accommodation and condensation coefficients (Fukuta and Walter, 1970). However, during the droplet growth and evaporation processes, the CCN collect inactive particles as CCN or as Aitken nuclei by the Brownian collision, diffusiophoresis, thermophoresis and collision due to the gravitational settling of droplets (Radke and Hobbs, 1969). This causes an essential change in the composition, size and activity of the CCN. Under the gravitational field, these processes proceed simultaneously and make it difficult to assess the extent of the contribution of each process.

In this sense, the memory effect of CCN is different from that of ice nuclei. However, as long as the CCN behavior alters, depending on their experience during the cloud formation and evaporation process, it must be clarified, for the process significantly contributes to the overall evolution of CCN in the atmosphere.

O.1.2 Objective

The objective of this experiment is twofold; first, to find out the change in activity of the CCN due to different cloud processes in terms of supersaturation and the duration using the room air, and second, to examine the effect of chemical and physical factors, such as hydrophobicity of insoluble particles and solubility, on the CCN activity after simulated cloud processes.

### O. 1.3 Scientific Justification

This study will clarify change in the behavior of CCN after cloud formation and evaporation processes. Although a part of this study concerns scavenging of non-CCN particles by formed cloud droplets, it specifically provides information about the activity change of the CCN instead of population change in Aitken nuclei.

### O. 1.4 Applications

The following are possible areas to which the result can be applied:

Estimation of the effect of non-precipitable cloud processes on the overall atmospheric CCN budget

Estimation of CCN evolution in fogs

Effect of steam condensation on scavenging of Aitken nuclei in polluting smoke.

### O. 1.5 Terrestrial Laboratory Limitations

In studying the memory effect, CCN have to be activated repeatedly. During the repeated activations, the sample air must be kept free from external interferences. However, when the CCN are activated in the terrestrial laboratories, the particle settling in the chamber creates a serious problem.

### O. 1.6 Zero-Gravity Opportunities

The low gravity condition in the Zero-Gravity Laboratory solves the problem of particle settling in the terrestrial laboratory mentioned above.

### O. 1.7 Approach

#### O. 1.7.1 General

As stated in the above sections, although it is possible to carry out a similar study, the possible range of study of CCN memory effect in terms of time, supersaturation and fog size is severely limited in the terrestrial laboratories due to the settling of formed cloud droplets in the memory test cycle. The Zero-Gravity Laboratory condition almost entirely removes such a restriction.

The CCN activation efficiencies will be studied within the continuous flow diffusion chamber after treatment of the sample air under various cycles of supersaturation, following undersaturation, and time durations in the expansion chamber.

Samples are the room air without modification, and smokes of hydrophobic, hydrophilic (water insoluble), and water soluble chemicals. For the hydrophobic smoke, dibutyl phthalate will be used. For the hydrophilic smoke, silicone dioxide (silica) will be used by decomposing a small amount of trisilane vapor in the air at high temperatures. Ammonium chloride and sodium chloride will be evaporated into water soluble smokes. Smokes of dibutyl phthalate and ammonium chloride will be generated by means of the simplified LaMer-Sinclair Monodispersed Aerosol Generator (see Class 13, Instrumentation). Decomposition of trisilane will be performed in the modified cooling tube of the smoke generator in which intense heating of the air-trisilane mixture is possible. The sodium chloride will be impregnated on a heating wire in the smoke generator and evaporated into smoke.

The experimental procedure is as follows. The smoke or air sample will be kept in the smoke chamber (see Class 13, Instrumentation). A small volume of the smoke sample will be taken out of the chamber and introduced into the expansion chamber which is at saturation conditions. When the sample smoke is well dispersed in the expansion chamber, a portion of the sample will be tested by the continuous flow diffusion chamber at several set supersaturation levels. Then, a slow adiabatic expansion will be made so as to stimulate the real supersaturation under cloud forming conditions. The supersaturation is normally on the order of 1 percent.

Strictly speaking, this real supersaturation is a function of expansion rates as well as the number of nuclei activated. Therefore, instead of trying to adjust the supersaturation level exactly to a predetermined level, the effect at the desired level of supersaturation will be estimated by interpolation out of a few experimental runs made around the level.

The uniform condition inside the expansion chamber does not last long in the terrestrial laboratories, due to the convective motion of air. Since this disturbance does not exist in the Zero-Gravity Laboratory, the expansion chamber should hold the uniform condition rather well except for the wall effect due to the molecular diffusion. The molecular diffusion however, is considered to be slow if the distance from the wall is larger than several centimeters, especially when fog droplets coexist. After having continued the expansion for a given period of time, the process will be reversed. The slow adiabatic compression will continue even after all the fog droplets evaporated until the relative humidity of the air inside the expansion chamber reaches to a fixed level. Then, the dry by memory carrying air will be introduced into the continuous flow diffusion chamber and measured at several set levels of supersaturation.

The same experimental procedure will be repeated for various rates of cooling and heating, time duration and relative humidity of drying. The ranges of the variables are:

Temperature of fog formation in the expansion chamber: room temperature and 5°C

Supersaturation

in Continuous Flow Diffusion Chamber: 0 - 2 percent

in Expansion Chamber: 0 - 2 percent

Relative humidity of drying in Expansion Chamber: 0 - 100 percent

Expansion Rate: 0.02 - 2 mb/sec ( $10^{-3}$  -  $10^{-1}$  °C/sec)

Duration for expansion and compression process: 1 - 10 min.

#### O.1.7.2 Instrumentation

A simplified LaMer-Sinclair Monodispersed Aerosol Generator will be used for smoke particle generation (the same as Class 13). This generator consists of a nuclei source (a heated Nichrome wire), evaporator of nuclei compound with dilution air inlet, mixing mechanism of the nuclei and vapor, and a cooling tube for gentle nucleation and growth. This generator uses a molten chemical to increase the vapor pressure. Use of woven fiber glass is recommended to restrain the liquid in order to prevent the liquid from floating.

For preparation of silicone dioxide smoke, a heating tube is necessary instead of a cooling tube. The trisilane vapor of known pressure will be mixed with air at a low but known ratio, and the gas mixture will be slowly passed through the heating tube. The Nichrome wire will be slowly passed through the heating tube. The Nichrome wire impregnated with NaCl will be used to generate the smoke.

The Continuous Flow Diffusion Chamber and the Expansion Chamber will be used. The Expansion Chamber must be capable of performing a slow expansion of the order 0.02 - 2 mb/sec. The wall temperature of the Expansion Chamber has to be controlled.

The smoke chamber to be used is about 1 m<sup>3</sup> in size with inlets for clean air and sample smoke, a port for sampling and an outlet to the smoke filter (the same as that in Class 13).

A multichannel recorder is required to record the temperatures of air and wall, and the pressure in the expansion chamber, the supersaturation and count of activated CCN in the Continuous Flow Diffusion Chamber.

### O. 1. 7. 3 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	
Type	3
Pollutant	
Pressure	
Temperature	10
Relative humidity	9
Charge	
Rate of cooling	3
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	



### O. 1.8 Procedure

General activity details are given below followed by a representative activity timeline. The event sequences and indicated times are based on knowledge of terrestrial requirements and restrictions with thought to translating this information to a low-g environment. Much more effort will be required to make these timelines operational, effective and efficient (see Figure O-1).

Activities		Minutes
•	Generate nuclei sample and store in smoke chamber	20
•	Activate CFD chamber	
•	Purge E chamber	5
•	Establish thermal and vapor equilibrium in E chamber	15
•	Inject samples into E chamber	
•	Introduce a portion of smoke into CFD chamber	
•	Record CCN counts at different supersaturations	10
•	Record wall and air temperatures in E chamber	
•	Expand E chamber slowly	10
•	Stop expansion and compress slowly to dry	10
•	Record relative humidity of E chamber	
•	Introduce E chamber smoke into CFD chamber	
•	Record CCN count and supersaturation	10
•	Recycle at other expansion-compression rate	
•	Recycle at other relative humidity of drying	
•	Recycle for other nuclei	

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Generate nuclei and introduce into smoke chamber</li> </ul>	20
<ul style="list-style-type: none"> <li>• Activate CFD chamber</li> </ul>	
<ul style="list-style-type: none"> <li>• Purge E chamber</li> </ul>	5
<ul style="list-style-type: none"> <li>• Establish thermal and vapor equilibrium in E</li> </ul>	15
<ul style="list-style-type: none"> <li>• Take smoke out of smoke chamber and inject into E chamber</li> </ul>	
<ul style="list-style-type: none"> <li>• Introduce a portion of smoke into CFD</li> </ul>	
<ul style="list-style-type: none"> <li>• Record CCN counts at 5 supersaturation levels</li> </ul>	10
<ul style="list-style-type: none"> <li>• Record wall and air temperatures in E chamber</li> </ul>	
<ul style="list-style-type: none"> <li>• Expand E chamber slowly</li> </ul>	10
<ul style="list-style-type: none"> <li>• Stop expansion and compress slowly to dry</li> </ul>	10
<ul style="list-style-type: none"> <li>• Record relative humidity of E chamber</li> </ul>	
<ul style="list-style-type: none"> <li>• Introduce dry smoke of E chamber into CFD</li> </ul>	
<ul style="list-style-type: none"> <li>• Record CCN counts at 5 supersaturation levels</li> </ul>	10
<ul style="list-style-type: none"> <li>• Recycle at other expansion-compression rates</li> </ul>	
<ul style="list-style-type: none"> <li>• Recycle at other relative humidities of drying</li> </ul>	
<ul style="list-style-type: none"> <li>• Recycle for other nuclei</li> </ul>	

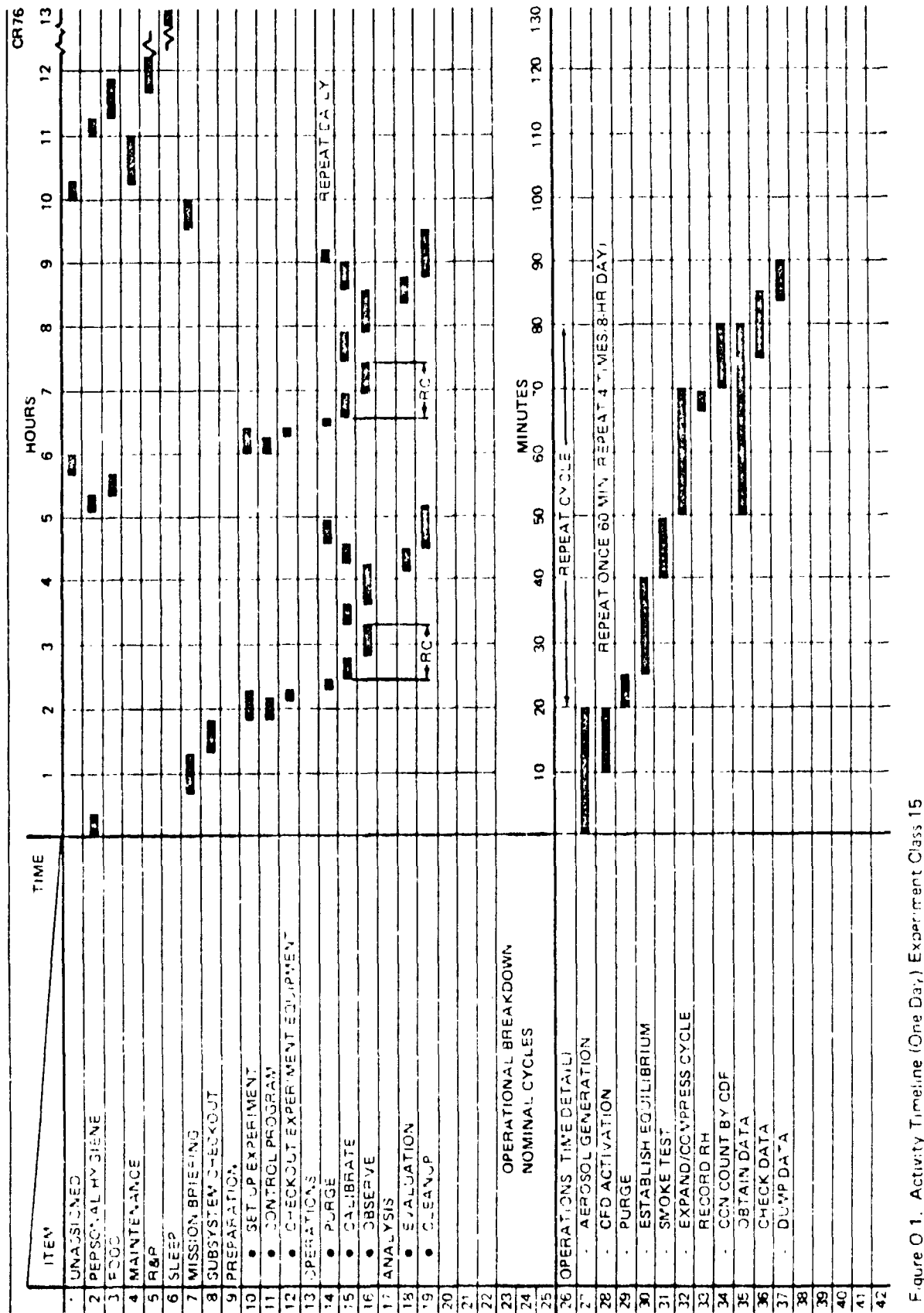


Figure O 1. Activity Timeline (One Day) Experiment Class 15

O. 1. 9 References

Fukuta, N., and L. A. Walter, 1970: Kinetics of hydrometer growth from a vapor-spherical model. J. Atmos. Sci., 27, 1160-1172.

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Smith, J. G., J. L. Kassner, Jr., and A. H. Biermann, 1968: Evaporation of small droplets in a Wilson Cloud chamber. Properties of residual nuclei. J. Rech. Atmos., 3, 41-44.

## O.2 CONDENSATION NUCLEI MEMORY APPROACH 2

### O.2.1 Introduction

Squires first focused attention on the fact that the differences in maritime and continental clouds could be accounted for by differences in the activity of the nuclei. In the succeeding years a great deal has been learned about the component of the natural aerosol which is effective in cloud formation processes; however, the interactions of different portions of the aerosol spectrum remains poorly understood. Some cloud condensation nuclei are believed to possess a memory effect. However, the existence, nature and relevance of the effect to microphysics is very poorly understood because few investigations have been attempted. In view of the potential that such a mechanism possesses for altering the spectrum of cloud condensation nuclei through processing in and out of cloud, this effect should receive more attention in controlled laboratory experiments.

### O.2.2 Objective

The objective of this experiment is to determine the effect of repeated cycling through cloud on the nature of the cloud condensation nuclei. It is postulated that during the time the nuclei are activated, the cloud drops possess from  $10^2$  to  $10^4$  times the efficiency as scavengers of nuclei in the smaller end of the spectrum. Moreover, the freshly created liquid surface may scavenge gaseous materials and serve as a site for chemical reactions. In addition the dissolving and recrystallization of the soluble salts will change the physical characteristics of the composite nucleus. This experiment is designed to test the hypothesis that the history of a cloud nucleus plays an important role in determining its activity as a cloud condensation nucleus.

### O.2.3 Scientific Justification

Squires first pointed out that the important differences between the colloidally stable continental clouds and the easily precipitating maritime clouds could be accounted for by differences in the activity and concentration of the cloud condensation nuclei. Our understanding of the nature of the condensation nucleus spectrum has advanced appreciably throughout the last two decades, although many questions remain unanswered. The metamorphosis of nuclei,

as a result of their processing through cloud, is important to the resolution of the question of the origin of the characteristic nucleus spectrum. The term memory effect has been carried over from general studies of the ease with which phase transitions are initiated in systems which have been subjected to repeated cycling. This is probably too specialized a definition to apply to the metamorphosis of cloud nuclei.

Rinker (1958), Dawborn (1965), Smith (1966), and Smith, Kassner and Biermann (1967) reported on the memory effect associated with the re-evaluation of droplets which had been formed by homogeneous nucleation, i. e., without the aid of any particulate nuclei. What they observed was residues resulting from impurities which become part of the droplet during the growth of the droplet but which did not redisperse upon evaporation of the droplets. Even though in the case of cloud nuclei the end result is a change in character of the phase transition process, the mechanisms involved in the alteration of the nuclei may be quite different from the memory effect associated with other phase transition processes. It is well known that interfaces tend to accumulate impurities.

A number of different processes can be envisioned as contributing to the metamorphosis of cloud nuclei. During the time when the nuclei reside in cloud as fully activated nuclei, soluble materials dissolve. Upon re-evaporation, the recrystallization of salts may alter the physical characteristics of the nucleus even to the point of shattering the solid material. When the nucleus is activated, the liquid surface may provide a site for chemical reaction with gaseous constituents of the atmosphere or the adsorption of dissolving of gaseous impurities (gaseous scavenging). Probably the most important factor involved in the metamorphosis of cloud nuclei results from in-cloud scavenging of particulates which have not been directly involved in the initial formation of the cloud droplets, since it is known that the latter effect is an important atmospheric cleaning mechanism. However, the effect of these processes on the metamorphosis of cloud nuclei has not been carefully studied, perhaps because facilities with which to simulate the slow and small changes in atmospheric thermodynamic conditions are not currently available. The difficulty in performing experiments of this nature in the terrestrial laboratory is compounded by the relatively high terminal velocity of cloud droplets which must be suspended for periods comparable to the lifetime of droplets in

natural cloud. Moreover, the shattering of crystalline nuclei upon drying may be strongly size dependent and dependent upon the presence of organic films which may or may not be effective in holding crystalline fragments together. In reality this experiment is closely related to several other experimental classes proposed for the Zero-G Cloud Physics Program and perhaps in the final analysis some of these other experiments can be modified to encompass portions of the needs of this experiment class.

It is well established that the marine environment does not possess the abundance of nuclei found in continental air. The earth-air interface of the continents is virtually a continuous source of condensation nuclei. Moreover, forest areas are rich sources of organic vapors which become involved in photochemical reactions. The blue haze which is characteristic of the Blue Ridge Mountains is aerosol which results from the photochemistry associated with the turpenses evolved from the forest of that area. The amazing thing is that the continental aerosol exhibits such a high degree of uniformity of activity that the initial diffusional cloud formation on this aerosol tends to produce a very narrow drop size distribution with uniformly higher drop concentrations than is found in marine clouds. If there were any appreciable number of superactive nuclei in the continental aerosol, one would expect them to behave in much the same way as giant salt nuclei behave in the maritime environment, i. e., they should appreciably broaden the drop size distribution.

One possible explanation for the uniformity of the continental aerosol is that the processing of nuclei through a cloud tends to uniformize their composition and size through the processes mentioned earlier. Thus the very process of condensation, evaporation and recondensation tends to make the aerosol more uniform in character. This could be an important step in developing a better understanding of the nature of the natural aerosol.

#### O.2.4 Applications

Evidence is beginning to accumulate that indicates that the natural aerosol possesses characteristics which are regional in character. For instance this might be one explanation for the widely differing values of sticking and thermal accommodation coefficients measured by various investigators at different locations. These may be strongly dependent upon the nature of trace

substances found in the local atmosphere. That portion of the continental aerosol which is involved in cloud formation is amazingly uniform in activity giving rise to clouds which are colloidally stable. The so-called cloud nucleus memory effect may be one of the mechanisms which tends to uniformize this portion of the natural aerosol. A more thorough understanding of this process is not only relevant to understanding the interaction of cloud with the atmospheric aerosol but it is also important in understanding how man-made pollutants might affect natural weather processes through inadvertent weather modification.

#### O.2.5 Terrestrial Laboratory Limitations

The principal processes which one can envision as being responsible for altering the characteristics of the nuclei which process in an out of cloud are: (a) trace gas scavenging, (b) particulate scavenging, and (c) alteration of the physical characteristics of the nucleus through dissolving and recrystallization of the soluble component of the materials (possibly including the break-up of the crystalline materials). The part of the aerosol spectrum which is likely to play the most important role in aerosol scavenging by the cloud droplets is that part which is just slightly smaller than the portion which is active as cloud nuclei. This part of the aerosol spectrum consists of relatively large particles with small diffusion coefficients. Therefore, in order to produce measurable results, the cloud and aerosol must interact for relatively long periods of time. The terminal velocity of cloud droplets is such that they fall out of the terrestrial chamber fairly quickly. Large chamber would not be expected to be suitable for such observations because it is impossible to guarantee uniformity in aerosol character throughout great expanses of chamber and in this type of experiment one is looking for small effects.

#### O.2.6 Zero-Gravity Opportunities

In scavenging experiments one needs a volume just large enough to minimize small particle diffusions to the walls but not so large that extremely uniform, well characterized aerosols cannot be easily supplied for the experiment. Gravity prevents the experimenter from realizing spatial homogeneity. As the cloud falls out under gravity the upper portions of the chamber are not subjected to scavenging for the same length of time as the lower portions of the chamber. When the cloud is evaporated, the various portions of the chamber



are likely to mix due to thermal inhomogeneities created by the latent heat released during evaporation. In Zero-G, these effects become nonexistent so that one possesses the ideal set of conditions for recycling nuclei in and out of cloud in an expansion type cloud chamber.

#### O.2.7 Quantification

In Zero-G one can hold a cloud in the cooled wall expansion chamber indefinitely and it will remain essentially homogeneous spatially with respect to droplet concentration and size distribution. This is what is required to provide a situation for gaseous and aerosol scavenging which can be subjected to unambiguous numerical modelling in order to analyze the data. In the terrestrial laboratory the continual differential settling of the cloud droplets produces monstrous problems in modelling the processes which take place in the chamber. Moreover, when the cloud is evaporated the inhomogeneities in cloud density produced by settling cause convection currents (arising from the latent heat absorption) which mix the system thereby averaging out much of the information which might have been obtained. At this stage in the development of the science, many years of effort will be saved by using the zero-g laboratory for such investigations. In the terrestrial case, when the number of undetermined coefficients or parameters becomes too large, so much information is lost that the intended results might only emerge as a vague qualitative understanding of the phenomena. Many body systems such as an aerosol are already enormously complicated. Experiments need to be kept as simple as possible to yield meaningful information. Zero-g provides these requirements.

#### O.2.8 Approach

##### O.2.8.1 General

General comments on the operation of an expansion chamber system can be found under the Approach I of Class 6—Scavenging.

The cooled wall expansion cloud chamber is to be utilized in its normal mode of operation. Cloud nuclei will be introduced in concentrations of around  $300 \text{ cm}^{-3}$ . Smaller nuclei will be introduced at much higher concentrations so that there is nearly a continuous spectrum from the cloud nuclei down into the Aitken nuclei. The nuclei spectrum will be characterized using both

thermal diffusion chambers and Aitken nucleus counters. The normal decay of the aerosol in the absence of cloud will have been studied in terrestrial laboratories but one examination of this decay may be in order for the zero-g experiment. A similar aerosol sample will be subjected to a predetermined simulated updraft velocity of about 1 mm/sec whereupon a cloud is formed with a drop concentration of about  $300 \text{ cm}^{-3}$ . The expansion may be stopped at this point if the mean size of the cloud droplets is to be kept small and it may be extended considerably if much larger mean drop size is desired. By varying the mean size and concentration, such parameters as the area presented by the surface of the droplets could be held nearly constant if desired. The experience gained with terrestrial facilities will be useful in writing detailed programs for the zero-g expansion chamber so that clouds can be formed to specification with a high degree of reproducibility. Another variable at our disposal is the simulated updraft velocity which regulates the rapidity with which the cloud develops; it need not be a constant even during a single experiment.

After the desired cloud properties have been obtained, the cloud growth can be terminated by stopping the expansion. The sensitive time of the cooled wall expansion chamber is so long that virtually no reheating occurs as indicated from experience with the UMR chamber. Thus, varying times should be allowed for scavenging to take place; a good guess that this time should range from 0 to 30 minutes in order to get the kind of data needed. The zero time is included so that the scavenging which takes place during cloud growth can be subtracted out since both diffusiophoretic forces and a continually changing droplet surface area will introduce extraneous effects not easily accounted for theoretically. The chamber will then be recompressed to its original state, returning the nuclei to their unactivated state.

At this point, two different forms of diagnostics should be used on alternate runs. First, the gas in the chamber can be analyzed with nuclei and particle counters in order to note changes in the characteristics of the nucleus spectrum. Second, the chamber can be re-expanded using exactly the same expansion program as before, this time noting any changes in the drop

concentrations and size distribution which might have resulted from any memory effect.

It is quite obvious that standard techniques are employed throughout this experiment with one possible exception. Some technique needs to be identified for measuring the drop size distribution in the chamber. The laser Doppler technique employed in the UMR terrestrial facility is not applicable to zero-g since it makes use of the terminal fall velocity of droplets under the influence of gravity. However, the drop size distribution could be sensed by optical light scattering counters by interrupting the experiment at the point where the size distribution is to be measured. No major obstacles are seen to be in the way of developing this experiment for the zero-g cloud physics laboratory.

The general procedure outlined above is based on this experimenters experience in examining memory effect with fast expansion cloud chambers, see Rinker (1958), Dawborn (1965), Smith (1966) and Smith, Kassner and Biermann (1967). By June 1975 the UMR cooled wall expansion chamber will be capable of handling some preliminary experiments relating to cloud nuclei memory effect which should comprise a useful feasibility study for the experiments outlined for the zero-g cloud physics program.

#### O.2.8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	3
Size-droplet	
Type	3
Pollutant	2
Pressure	
Temperature	6
Relative humidity	6
Charge	
Rate of cooling	2
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### O.2.9 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (See Figure O-2).

#### O.2.10 References

1. Smith, J. G., Kassner, J. L., Jr., and Biermann, H. H. (1968), "Evaporation of Small Droplets in a Wilson Cloud Chamber, Properties of Residual Nuclei," J. de Recherches Atmospherique 3, 41.
2. Smith, J. G., (1966), "Re-evaporation Nuclei and Evaporation in a Wilson Cloud Chamber, " Ph. D Dissertation, University of Missouri-Rolla.
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# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on all electrical components	5
• Check operational readiness of components	5
• Check and calibrate control and measuring circuits	10
• Turn on condensible gas supply and sump pump	0.5
• Open expansion chamber ports	0.5
• Purge system	10
• Load control program into control computer	(5)
• Load time lapse camera	(3)
• Close expansion chamber ports	0.5
• Turn on thermal controls: conditioning chamber, humidifier, expansion chamber	1
• Adjust gas flow to humidifier	0.5
• Adjust gas flow to aerosol generator	0.5
• Turn on aerosol generator and pollution gases	1
• Turn on hygrometer, particle mass meter, particle size spectrometer	1
• Sample relative humidity, nuclei size, mass, concentration	10
• Adjust humidifier and aerosol generator as required	(-)
• Open expansion chamber ports	0.5
• Transfer monitoring points to output of expansion chamber	0.5
• Monitor sample relative humidity, nuclei size, mass concentration	8
• Close expansion chamber ports	0.5
• Turn on meter, expansion mechanism, pressure control, gas temperature, video, laser, liquid water meter, drop size	1
• Let expansion chamber come to thermal equilibrium	10

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn off aerosol generator, thermal controls for humidifier and conditioning chamber, particle size, mass and concentration meters, hygrometer	(2)
• Purge system (except expansion chamber)	(4)
• Turn off noncondensable gas supply and sump pump	(1)
• Turn on time-lapse camera	0.5
• Start cooling/heating program	30
• Observe chamber on video monitor	(-)
• Stop time-lapse camera	0.5
• Turn off pressure control, expansion mechanism, video, laser, liquid water meter, drop size, meter	1
• Warm chamber to +10°C and shut off expansion chamber thermal control	5
• Open expansion chamber ports	0.5
• Turn on noncondensable gas supply and sump pump	0.5
• Purge chamber	8
• Unload time-lapse camera and store film	(3)
• Check computer data	(4)
• Transfer data to permanent storage	(1)
• Close expansion chamber ports	
• Turn off noncondensable gas supply and sump pump	0.5
• Shut down system or start next experiment	10

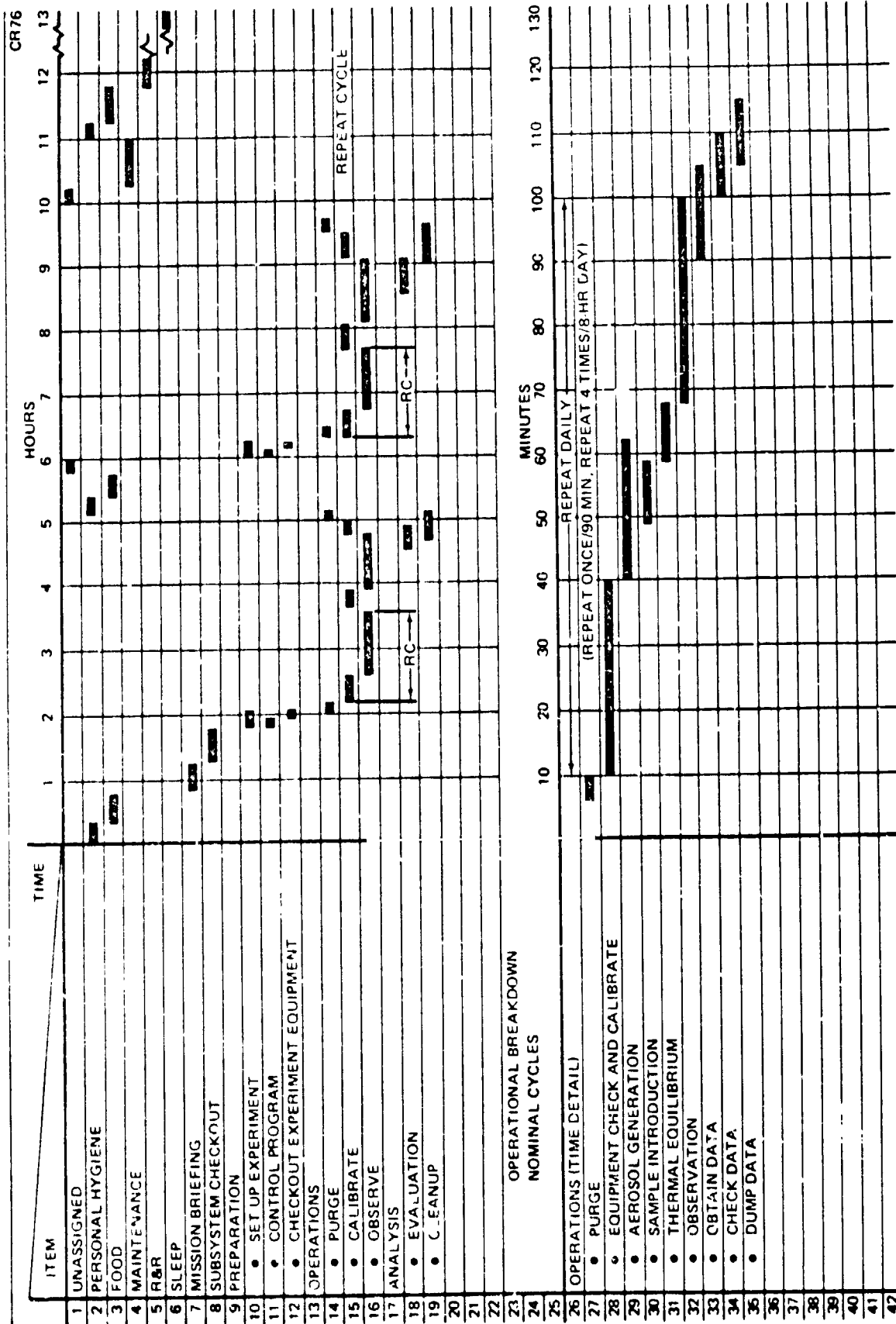


Figure O-2. Activity Timeline (One Day) Experiment Class 15



Appendix P  
CLASS 16  
NUCLEI MULTIPLICATION

P.1 NUCLEI MULTIPLICATION APPROACH 1

P.1.1 Introduction

The importance of sea salt particles as cloud condensation nuclei has long been recognized. Much work has been done in attempts to correlate the nature of marine aerosol with processes taking place at the sea surface. One of the pressing problems is how salt particles become subdivided into the smaller size ranges. A number of investigators have noticed that brine droplets which dry out and crystallize very often tend to shatter into a few larger nuclei and a host of very small particles. Such a process is important to understanding the size distribution of salt particles in the atmosphere.

P.1.2 Objective

The specific purpose of the experiment is to investigate: 1) the growth and evaporation of a population of giant salt nuclei in a chamber simulating their processing in and out of clouds for humidity cycles extending from several minutes to several hours, 2) observation of the structural changes including possible shattering of those nuclei during the humidity cycles, and 3) the growth of a population of salt particles mixed with inactive aerosols and subjected to the passivation by surfactants in humidity cycles.

P.1.3 Scientific Justification

The importance of sea salt particles as condensation nuclei has been recognized by Kohler (1926), Findeisen (1939), Simpson (1941), Mason (1957), and Woodcock (1950) in the first half of this century. Usually their existence was connected with the starting of a condensation process inside of an air mass with high relative humidity. However, G. C. Simpson (1941) expressed an

idea that giant hygroscopic nuclei might start a coagulation process as well. Findeisen (1939) observed during World War II an unusual development of precipitation process over the North Atlantic (Schulz, 1947) which led to a light rain from clouds having relatively small thickness and their tops not surpassing the zero degree isotherm. The idea of the initiation of precipitation by giant salt nuclei was supported by Woodcock (1952), who found sea salt particles larger than  $10\text{ }\mu\text{m}$  in the maritime atmosphere, and by theoretical studies of the development of the precipitation process within pure water clouds by Bowen (1950), Ludlam (1951), Mason (1952), Shishkin (1951, 1952), and by Telford (1955).

The whole precipitation stimulation process by salt nuclei can be divided into two parts: the condensational growth of embryos as supersaturation develops and the stage of coagulation of the few large drops (grown on giant nuclei) with the tiny cloud elements.

Old measurements and observations by Wall (1942) showed that hygroscopic particles exposed to increasing humidity adsorb a few molecular layers of moisture at low relative humidity. Wall's observations, however, were made with bulk materials. Unlike Wall, Orr et al (1958) paid attention to the change of the size of small hygroscopic aerosol particles with increasing relative humidity. These authors found an insignificant increase in size of sodium chloride particles of  $0.05\text{ }\mu\text{m}$  in diameter at 30 percent relative humidity; a volume of 2 percent was observed due to adsorbed water. However, this conclusion might be substantially changed if one assumes that the giant and large nuclei are often formed of aggregates of tiny salt particles. The last hypothesis is substantiated by recent studies of the shapes of giant salt nuclei on the seashore. These studies showed that the larger particles were composed of a large number of tiny salt crystals (Podzimek, 1974). The study of the behavior of giant salt nuclei at higher relative humidities seems to be one of the most challenging problems in cloud physics.

There are two different spheres of questions which might be important for the establishment of a model of growing embryos and water drops in a warm cloud. How are the giant salt nuclei transformed during periodical changes of the relative humidity and what structural changes occur in polycrystalline nuclei in the environment characterized by the presence of other chemical substances and by variable humidity.

The periodic changes of relative humidity will certainly cause an effect called in sea nuclei physics "memory effect" which in the case of cloud physical studies is closely related to the so-called "recycling of nuclei". Some authors have mentioned the "filtration" effect of clouds in which the majority of giant nuclei are retained in clouds and only a small fraction amount will be re-emitted out of the clouds (Podzimek, 1959). The emitted nuclei originate from evaporated cloud drops and are subjected to structural changes and probably are subject to nuclei shattering as the salts recrystallize.

H. Dessens (1946, 1949) observed shattering of sea salt particles in the samples collected on the seashore. Facy (1951) found a similar phenomenon in the laboratory at a relative humidity less than 70 percent. Later Twomey et al (1955) observed that at lower relative humidity each saline droplet produced several hundred minute particles in the size range between 0.01 and 0.1  $\mu\text{m}$ . Unlike Blanchard et al (1964), who could not find any evidence that salt particles would be generated in large numbers during salt solution droplet crystallization, Radke et al (1972) supported the hypothesis of nuclei shattering. Slightly supersaturated air containing salt particles yielded much higher concentrations of tiny nuclei than the original sample (multiplication factor of five times in the size range  $d < 0.062 \mu\text{m}$ ) after the nuclei passed through a heated tube. Podzimek and Saad (1974) pointed out the importance of the temperature of the heated drift tube for shattering effect. However, their multiplication factors were lower than those published by Radke et al (for drift tube temperatures slightly below 100°C, multiplication factors of approximately 2.5 were observed). Matijevic et al (1963) made a very interesting observation on the transformation of spherical sodium chloride

particles (generated by the furnace technique at approximately 900°C) into cubically shaped crystals after introducing them into a container with higher relative humidity. Similar studies support the importance of transformation of the structure of salt nuclei which might lead to the change of their nucleation activity.

The presence of other salts, inactive aerosol particles, and organic substances might also influence the activity of pure salt nuclei. Winkler (1973) compared the activity of pure salt nuclei with those found in the atmosphere. Atmospheric aerosol shows continuous smooth growth with increasing humidity, unlike pure salt particles which are characterized by a sharp increase in size at a critical value of humidity. The gradually increasing sizes of atmospheric aerosol particles are mainly due to the mixed nature of the nuclei. Insoluble substances were found in natural nuclei in addition to other hygroscopic compounds and organic material. Winkler found an increase of solubility of various ions due to their mutual interaction comparing it with pure salt particles. This is due to the effect of the solubility product which is known to chemists.

The passivation of sodium chloride condensation nuclei by some organic substances, mainly surfactants, was thoroughly investigated recently by Russian scientists. Bakhanova and Deryagin (1972), Leonov et al (1973), Chalenko et al (1973), and Morachevski and his fellow-workers (1968) studied mainly the retarded growth of drops of salt solution covered by cetyl alcohol. Marked effects were found mainly in the case of very small solution drops ( $r = 2 \mu\text{m}$ ) which supported Blanchard's (1969) findings that highly concentrated organic substances showed a much larger influence on the smallest drops in the maritime atmosphere. From the available literature on the influence of organic substances on the evaporation rate of drops of salt solution, we can conclude that there is a marked effect on the phase transition and on the formation of salt crystals as potential condensation nuclei.

All the known studies with giant salt particles were made with aerosols stored in relatively small containers. This arrangement excluded immediately the application of the results of similar studies for the simulation of large scale phenomena occurring in the atmosphere. There, the nuclei are subjected to fluctuations of humidity and temperature of the order of tens of minutes or hours. During this time the giant nuclei of principal interest would have settled out in the experimental simulation chamber.

#### P.1.4 Applications

Reasons for this proposal are apparent from the Introduction. The study of the precipitation from warm clouds is important in tropical and subtropical regions and to a limited extent in the maritime atmosphere in the middle latitudes. Giant sea salt nuclei apparently play an important role in the initiation of the precipitation process because they provide the breadth needed in the drop size distribution to get the collision coalescence mechanism going without undue delay. Their transformation and possible multiplication during the transport from the sea surface under changing relative humidity and temperature will influence the final aerosol size distribution and might strongly influence cloud condensation at the cloud base and the later development of precipitation elements. Several studies indicated the importance of inactive particles, of pollutants, and of surface active materials on the structural changes and activity of sea salt nuclei. We hope also to study in this way the possibility of artificially interfering in the cloud formation and in the subsequent development of the precipitation process.

The sea covers a large fraction of the earth's surface. Nuclei originating at the sea surface change in form by processes such as the nuclei multiplication mechanism envisioned in this experiment to yield the characteristic marine aerosol which determines the character of maritime clouds. This investigation has immediate application to understanding the origin and fate of the maritime aerosol. The marine aerosol also determines the character of the shoreline haze and coastal and harbor fogs. The break up of the salt aerosols is also of importance in understanding the ecological hazards of applying salt for the melting of ice and snow on highways and of using brine in power plant cooling towers.

#### P.1.5 Terrestrial Laboratory Limitations

Salt nuclei multiplication processes are likely to be predominantly confined to the larger nuclei, such as giant sea salt nuclei. In order to study this process, either salt solution droplets must be injected into the chamber and then evaporated or large well-characterized nuclei must be subjected to cloud formation and then subsequently evaporated under different types of schemes. Since the nuclei are large and the cloud droplets (whether they be spray droplets or cloud droplets grown on large hygroscopic nuclei) have relatively large terminal velocities, the total height needed in a terrestrial laboratory cloud chamber is of the order of 300 to 600 cm in order to provide ample time for the matriculation of the experiment. As the chamber volume becomes large, the difficulty of supplying a homogeneous sample throughout the volume becomes greater and the logistics of providing thermal uniformity throughout the chamber becomes greater. Although in principle, both of these problems can be handled in the terrestrial laboratory with the expenditure of suitable sums of money, other problems associated with the settling of the cloud into the lower parts of the chamber cause an inhomogeneity which cannot easily be overcome. Both the vapor and some of the active aerosol particles are shifted to lower regions of the chamber. After evaporation of the droplets, the vapor distribution is rendered inhomogeneous and bulk vapor diffusion then will tend to redistribute the vapor. This causes still another instability brought about by the fact that latent heat is carried from one part of the chamber to another which upsets the thermal balance of the chamber. What this means is that some very difficult numerical modeling must be undertaken in order to unravel the interaction of different processes inside the terrestrial chamber. In zero gravity, the whole cloud chamber volume would retain spatial uniformity in its vapor density, temperature, and nuclei concentration which would greatly add to the certainty with which an already complex problem can be interpreted. Some of the parameters, which would be needed in order to adequately model the processes in the terrestrial chamber, are candidate subjects for other investigations in the proposed zero gravity program.

#### P.1.6 Zero-Gravity Opportunities

Some of the proposed experiments can be performed in the terrestrial laboratory. However, we cannot use "ultragiant" salt nuclei due to their large settling rate and we cannot expect the high degree of homogeneity of temperature and humidity fields. We anticipate that zero-gravity conditions will enable us to perform experiments in durations of several hours.

#### P.1.7 Quantification

To our knowledge, the proposed experiments with giant nuclei can be done with less accuracy and only with nuclei suspended on a thin wire or with nuclei deposited on a substrate. Both approaches have several drawbacks related to the distortion of the results by the contact of solution drops with solid bodies. We are also excluding the random motion of particles important for the diffusional interaction of particles.

#### P.1.8 Approach

##### P.1.8.1 General

The proposed work assumes the use of an expansion chamber or, in general, a chamber permitting simulation of different humidities from R.H. = 30 percent to slight supersaturation at positive temperature. The instrumentation necessary for this program should enable generation of aerosol particles of wanted size, measurement of the size spectrum distribution as a function of time, and introduction of inactive particles and surfactants in a controlled amount and/or size distribution.

The proposal is based on the following results of preliminary studies. An extensive study has been done on different techniques of generation of sodium chloride giant nuclei within the size range from 0.1  $\mu$ m to several microns. From bubbling the air through a salt solution, spraying of salt solution, dispersion of salt solution drops in an ultra-acoustic mist generator, and from the evaporation of sodium chloride in a furnace at a temperature of 880°C,

we obtained the following conclusions. The most promising device seems to be the ultra-acoustic mist generator possibly combined with furnace techniques. It yields particles in the size range between tenths of microns and several microns in high concentration and with a sufficient degree of monodispersity (dispersity factors  $\alpha = 0.30$  to  $0.40$ ). The method can be refined for larger sizes of particles.

The study of the changes of stored sodium chloride particles in a mylar bag with the aid of a mass monitor, Royco and Aitken nuclei counter, and multichannel analyzer reveals that, after the initial period of a strong decrease in concentration of large giant nuclei ( $d > 5 \mu\text{m}$ ) and the decrease of smaller nuclei according to the coagulation equation, a sudden increase of particles with diameters  $1.4$ ,  $0.7$ , and  $0.5 \mu\text{m}$  was observed after 45 to 90 minutes. This might be explained assuming the shattering of larger particles. The analysis of electromicrophotographs supports this hypothesis (Podzimek, Saad, 1974).

A preliminary theoretical study of the possible meteorological effect of the increasing concentration of giant and large nuclei as well as on the colloidal stability of the cloud base has been discussed and the primary importance of the numbers of larger giant nuclei was found (Podzimek, Saad, 1974). There is a strong indication that the presence of surfactants can influence considerably the development of a warm cloud.

To determine the extent of nuclei multiplication occurring when a drop containing a soluble material evaporates, droplets of the desired size and solution concentration will be generated and placed in the expansion chamber. The initial pressure, temperature, and relative humidity will be chosen such that the droplets can be evaporated using an adiabatic compression of the chamber.



The initial concentration of droplets will be determined from photographs taken by the time lapse camera (holography, if it is available). After the droplets have been evaporated and the soluble material crystallized, an adiabatic expansion will be used to activate the nuclei present. If there has been a multiplication of nuclei due to break up of the nuclei during crystallization, the concentration of nuclei observed during the search expansion will have increased. The number of nuclei activated as a function of time, temperature, and supersaturation during the search expansion will also provide some information as to the distribution in supersaturation required to activate the new nuclei.

An alternate approach would be to introduce condensation nuclei of the soluble material in the chamber and form the droplets in the chamber by an adiabatic expansion. The droplets would then be evaporated by an adiabatic compression after recording the initial concentration. The compression would then be followed by the search expansion.

#### P.1.8.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	3
Type	9
Pollutant	3
Pressure	
Temperature	3
Relative humidity	3
Charge	
Rate of cooling	
Time	2
Sound	
Electric field	
Nuclear radiation	
Adsorption	

C-5

<u>Parameters</u>	<u>Variations</u>
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### P.1.9 Procedure

General activity details are given below (Figure P-1) followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient.

The system is first turned on and all components checked for operational readiness. The measurement and control circuits are checked and calibrated if necessary. The ports of the expansion chamber are opened and the system purged with clean air. While the system is being purged, the control program and operational parameters of the intended experiment are loaded into the control computer. When the purging has been completed, the ports of the expansion chamber are closed and the air flow to the humidifier and drop generator adjusted to the required values. The sample is monitored as

it leaves the conditioning chamber for drop size and concentration and relative humidity. When the sample stabilizes, the humidifier temperature is adjusted to give the desired relative humidity and the drop generator to give the correct size and concentration of drops.

When a stable usable sample is achieved, the ports of the expansion chamber are opened and the chamber flushed with the sample. Flushing continues until the sample leaving the chamber has stabilized. The expansion chamber ports are then closed and the chamber allowed to come to thermal equilibrium. While the chamber is coming to thermal equilibrium, the drop generator is turned off and the rest of the system purged with clean air.

When the expansion chamber has come to thermal equilibrium, the time lapse camera is started. The chamber is then compressed and heated to evaporate the drops. After the drops have evaporated and the solute crystalized, the chamber is expanded to produce drops on the nuclei present. During this period of evaporation and condensation, the drop size and liquid water content are recorded.

After the expansion has been completed, the time lapse camera is turned off, unloaded, and the film stored for later processing. The computer data is checked for any gross malfunctions and the data then transferred to permanent storage for later detail analysis. The expansion chamber ports are opened and the chamber purged with clean air. When the expansion chamber is clean, the ports are closed again and the entire system shut down.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Turn on all electrical components.	5
● Check operational readiness of components.	5
● Check and calibrate control and measuring circuits.	10
● Turn on noncondensable gas supply and sump pump.	0.5
● Open expansion chamber ports.	0.5
● Purge system.	10
● Load control program into control computer.	(5)
● Load time-lapse camera.	(3)
● Close expansion chamber ports.	0.5
● Turn on thermal controls: conditioning chamber, humidifier, expansion chamber.	1
● Adjust gas flow to humidifier.	0.5
● Adjust gas flow to drop generator.	0.5
● Turn on drop generator and pollution gases.	1
● Turn on hygrometer, drop mass meter, and drop size spectrometer.	1
● Sample relative humidity, drop size, mass, concentration.	10
● Adjust humidifier and drop generator as required.	(-)
● Open expansion chamber ports.	0.5
● Transfer monitoring to output of expansion chamber.	0.5
● Monitor sample: relative humidity, drop size, mass, concentration.	8
● Close expansion chamber ports.	0.5
● Turn on expansion mechanism, pressure control, gas temperature video, laser, liquid water meter, drop size meter.	1

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Let expansion chamber come to thermal equilibrium.	10
● Turn off drop generator; thermal controls for humidifier and conditioning chamber; drop size, mass, and concentration meters; hygrometer.	(2)
● Purge system (except expansion chamber).	(4)
● Turn off noncondensable gas supply and sump pump.	(1)
● Turn on time-lapse camera.	0.5
● Start cooling/heating program.	30
● Observe chamber on video monitor.	(-)
● Stop time-lapse camera.	0.5
● Turn off pressure control, expansion mechanism, video, laser, gas temperature, liquid water meter, drop size meter.	1
● Heat chamber to +10°C. Turn off expansion chamber thermal control.	5
● Open expansion chamber ports.	0.5
● Turn on sump pump and noncondensable gas supply.	0.5
● Purge chamber.	8
● Unload time-lapse camera and store film.	(3)
● Check computer data.	(4)
● Transfer data to permanent storage.	(1)
● Close expansion chamber ports.	0.5
● Turn off noncondensable gas supply and sump pump.	0.5
● Shut down system or start next experiment.	10

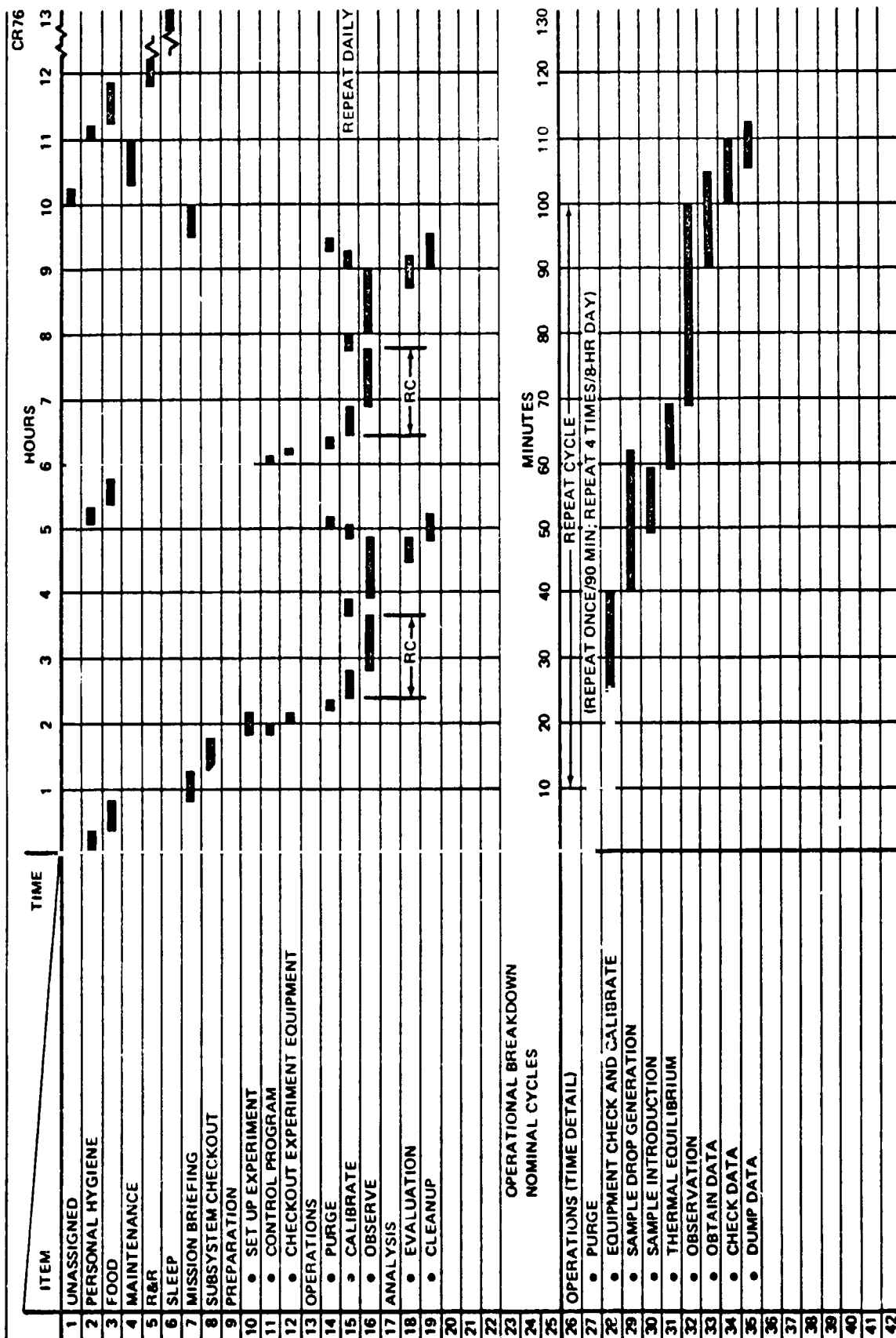


Figure P-1. Activity Timeline (One Day) Experiment Class 16

#### P.1.10 Mission Timeline

Until there is more detailed knowledge of the techniques that will be used in the final version, the activity timeline for the zero-g experimental procedure is the same as that developed for the terrestrial experiment (see Figure P-1).

It is possible that purge times and rates can be reduced but this can be determined only by testing of the actual equipment. The degree of automated control will effect the time required for some operations in both the zero-g and terrestrial experiments.

The primary change in the time line for zero-g operation will be an extension of the duration of the expansion/contraction cycle to make full use of the zero-g environment.

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## P.2 NUCLEI MULTIPLICATION APPROACH 2

### P.2.1 Introduction

The precipitation processes are a function of available nuclei size and number. Large ( $0.1\text{ }\mu\text{m}$  to  $1\text{ }\mu\text{m}$ ) and giant ( $1\text{ }\mu\text{m}$  to  $10\text{ }\mu\text{m}$ ) NaCl nuclei in particular play a major role. The oceans are the main source of salt nuclei which are produced as a result of the formation and subsequent evaporation of droplets formed by the breakup of waves and bubbles at the ocean surface. The large NaCl particles are known to exist at much lower concentrations over land masses than over the oceans. A number of processes, including the particle breakup during evaporation, are believed to contribute to this decrease. Knowledge of the depletion processes causing this loss of large NaCl particles would provide a link to the understanding of the nuclei size and mass distribution in the atmosphere. Better understanding of this breakup mechanism could also play an important role in the weather modification technique of dispersing NaCl where precise size and particle numbers are required. Salt particles are important nucleating agents for oceanic and shoreline haze problems. Better understanding of the breakup process would lead to improved forecasting capabilities and to eventual haze and fog modification and control techniques.

Brine cooling towers are considered a method of avoiding the thermal pollution of lakes and rivers during the generation of electrical power. One aspect of brine towers is the significant loss of the saturated solution to the ambient air. The rate of accumulation of the salt from the brine depends on droplet size and fall velocity. Present theoretical considerations, neglecting particle breakup, indicate that an undesirable salt accumulation could occur in an area around the towers. If salt particle breakup existed during the rapid evaporation of the brine droplets, the salt would be dispersed over a greater area. The concentration accumulation decreases by as much as the fourth power of the particle diameter. Thus, if the particle diameter decreased by a factor of 2, the concentration would fall by a factor between 4 and 16 depending on particle size. Thus, the ecological impact depends on the dispersion processes and determines the non-use, use, and design of towers versus other cooling methods.

There has been concern about the damage done by salt wash-off from highways. Another aspect of this problem is the generation of salt mist due to vehicle motion over salt laden highways. The distance that this salt mist disperses depends on salt size and numbers. Salt particle breakup during the evaporation of the salt droplets would be important in the determination of the ecological impact of the use of salt on highways.

#### P. 2. 2 Objective

The objective of this experiment is to determine the processes and extent of nuclei material breakup.

#### P. 2. 3 Scientific Justification

The following reiterates the conceptual model of possible nucleus multiplication. Possibly it occurs when the evaporation rate is rapid compared with the internal mixing rate of the droplet. This results in the formation of a solid shell of solute at the drop surface which must be ruptured for further evaporation to occur. This rupture could well lead to a hollow bead structure similar to freeze-dried coffee, and the aerosolization of the liquid in the interior of the drop. This in turn should cause a sudden acceleration of the drop which would be clearly detectable under completely still air and zero-gravity conditions. Even a photographic arrangement that did not completely resolve the details should be capable of detecting this acceleration, and perhaps the spray of minute droplets from the inside. The critical conditions for this to occur would then be a function of temperature, humidity, and droplet size (or rather, the size of a droplet that was just saturated, assuming that the initial droplet might or might not be a saturated solution). The use of several droplet sizes and humidities should permit homing in on the sorts of combinations of size and evaporation rate that are necessary for the phenomenon to occur. This could be affected in either direction by the presence of impurities other than the principal solute (presumably sodium chloride), and it is therefore proposed that the more realistic sea water be used as well as pure salt. Five replications of each set of conditions will be used so as to obtain some statistical validity for whatever is observed.

#### P.2.4 Applications

The extent of nuclei breakup is important in the shaping of the nuclei size distribution in the atmosphere. Salt particles, e. g., are an important nucleating agent in oceanic and shoreline haze problems. Better understanding of the breakup process and conditions could lead to improved haze forecasting and to eventual haze and fog modification and control techniques. This breakup also has ecological importance in relation to brine cooling towers and highway salting.

#### P.2.5 Zero-Gravity Opportunities

Present investigation of this important research problem has reached a plateau because the Earth's gravitational field prevents the observation of this sea-salt breakup process. Small particles are lost when vertical wind tunnels are used to investigate this phenomena. Mechanical supports modify the heat, electrical, and vapor processes and thus do not provide realistic answers. Even with mechanical support, small particles are lost due to gravity-induced "fallout." Thus, a low-gravity environment provides the time to study the primary particle and resulting smaller particles.

#### P.2.6 Approach

##### P.2.6.1 General

A given size of droplet with the nuclei material in solution (e. g., NaCl or ocean water) will be inserted into an expansion chamber of specified relative humidity below 80 percent. For low enough humidities, the droplet will evaporate and the nuclei material will crystallize. It is during the crystallization, which is believed to be very rapid, that the number of very small fragments may break away from the main particle. After crystallization, the appropriate expansion will cool the chamber giving a supersaturation which will result in the nucleation and growth of any small nuclei that were generated. Nuclei counts will be obtained by photography.

An alternate approach after breakup would be to pass the air through a continuous flow diffusion chamber for nuclei growth and then into an optical counter to provide information on size as well as numbers.

An early opportunity version of this experiment depends on photography and Nuclepore filters to provide the qualitative numbers for this type of experiment.

#### P.2.6.2 Chamber Subsystem

An analysis indicates that a chamber with total internal dimensions of 30 cm diameter by 45 cm high will be sufficient for a duration of several minutes, assuming acceleration values of  $<10^{-3}$  g as determined from Apollo 14 demonstration experiments. This chamber will have the following:

- A. Purge inlet and outlet.
- B. Interior blackened to minimize scattered light.
- C. Observation windows on two opposite walls.
- D. Droplet injection mechanism
- E. Sensors for temperature, pressure, and relative humidity.

#### P.2.6.3 Purge Subsystem

The purge subsystem is used to remove unwanted salt particles from the chamber and to control the RH of the chamber (Figure P-2). Two three-way valves will be used to control flow:

- Position a. Air Bypass.** This is used to filter out the salt particles from the chamber, but leaves the relative humidity unchanged.
- Position b. Dryer.** The total water vapor in the chamber at 20°C, 80 percent relative humidity is  $2.1 \times 10^{-4}$  gm. Assuming twice this value to include the purge system gives  $4.2 \times 10^{-4}$  gm of water. A desiccant such as a molecular sieve can absorb 0.2 gm of water per gram of its own weight and the sieve material has a packing density of 0.6 gm/cm<sup>3</sup>. These values indicate that 3 cm<sup>3</sup> of molecular sieve would be needed to completely dry the system. One hundred cm<sup>3</sup> of desiccant is a reasonable value which permits the chamber subsystem to be purged a number of times.
- Position c. Humidifier.** The humidifier would utilize surface tension and capillary action to maintain a moist outer surface of a ceramic tube. For proper humidification the air passage should not be greater than 2 mm, with enough length for several seconds of air residence time. The concept is shown in Figure P-3.

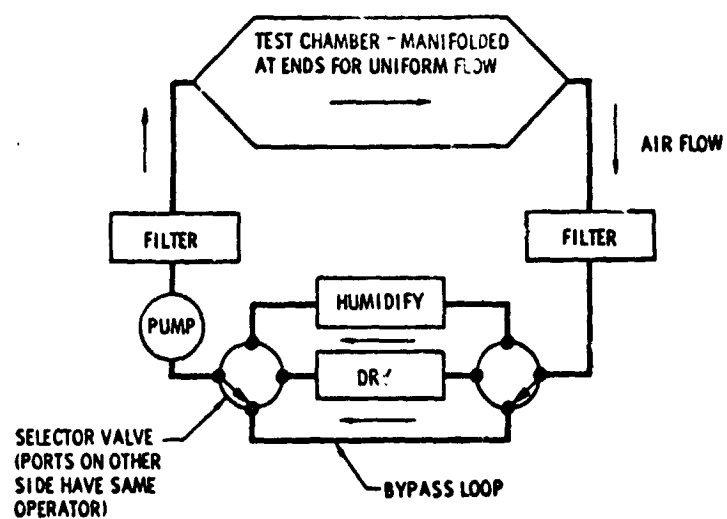


Figure P-2. Purge System Schematic

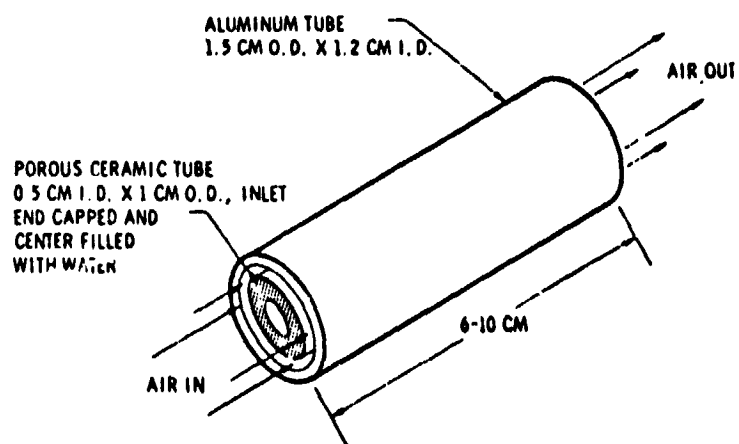


Figure P-3. Humidifier

Position d. The addition of a venting/intake valve will permit the necessary expansion and compression.

Pre- and post- 1  $\mu$ m absolute filters (e. g., Nuclepore filter) will be used to prevent salt particles from entering the humidity control section and also to prevent particles from entering the chamber.

An alternate chamber would be the expansion chamber, provided it could be modified to permit the introduction of single droplets of controlled size. It would have to be demonstrated that droplet introduction, did not produce extraneous nuclei, and that the presence of the necessary equipment did not result in nuclei being formed. If this condition can be fulfilled, then it is possible to do the experiment by expanding the chamber air repeatedly probably during a maneuver that produces some gravity, to get all nuclei out of it; introducing the drop, then causing it to evaporate by adiabatic compression, then expanding it to see whether only a single nucleus, or many nuclei, result. This provides less information about mechanism, but is probably more sensitive than the previous proposal, if you assume that multiplication is caused by some mechanism other than suggested above. The main difficulty is that chamber clean-up by the usual procedure of repeated expansion and subsequent rainout, does not occur under zero-gravity conditions. This is going to be a fairly constant problem with the expansion chamber. Possibly what will be necessary is a very careful control of the degree of expansion to avoid high super-saturations. Under these conditions, a Millipore or Nuclepore filter - or for that matter a tube properly packed with cotton wool - would probably do the job. Once again, of course, this is predicated on keeping the super-saturation small so that one did not have to worry about nucleation on ions and similar very small particles.

As a final point, nearly all of these designs are predicated on some assumptions concerning the behavior of cloud chambers under zero-G. It is not at all clear that these assumptions are valid. There are many possible phenomena that cannot be observed in a chamber under gravity, and that are perhaps too slow in their action to be observed in the drop tower. For example, refer to the work of Mason and his colleagues on the simulation

of falling bodies of various shapes in finite chambers. Because of wake interaction, these tended to arrange themselves in regular polygons. We may find that, no matter where drops are introduced into a chamber, they migrate to the precise center if they are alone, that four drops arrange themselves in a tetrahedron, or conversely that they are attracted to the walls and never stay any place at all. Until we have a real opportunity to observe a drop in a chamber over quite a long period of time, all of these experimental designs are inevitably going to be somewhat speculative. If this were not the case, the experiments would hardly be worth doing. There is certainly going to be no substitute for an observer who is really alert to observe what happens, to feed it back to the scientists at the ground, and to make improvisations as they are made necessary by unexpected phenomena.

#### P.2.6.4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	4
Type	
Pollutant	3
Pressure	4
Temperature	5
Relative humidity	5
Charge	3
Rate of cooling	
Time	
Sound	
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	



Parameters	Variations
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	4
Ion Level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### **P.2.7 Procedure**

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure P-4).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Purge chamber</li> <li>• Establish temperature, pressure and relative humidity</li> <li>• Insert solution droplet(s)</li> <li>• Photograph evaporation rate and motion</li> <li>• Expand to supersaturation</li> <li>• Photograph resulting droplets</li> <li>• Recompress-evaporate droplets</li> <li>• Expand photograph droplets</li> <li>• Recycle with more droplets of the same size (15 times)</li> <li>• Recycle for other droplet sizes (4 sizes)</li> <li>• Recycle for other humidity values (4 values)</li> <li>• Recycle for other temperatures (4 values)</li> <li>• Recycle for other pressures (3 values)</li> </ul>	<ul style="list-style-type: none"> <li>5</li> <li>10</li> <li>1</li> <li>2</li> <li>1</li> <li></li> <li>2</li> <li>1</li> <li></li> <li></li> <li></li> <li></li> <li></li> </ul>

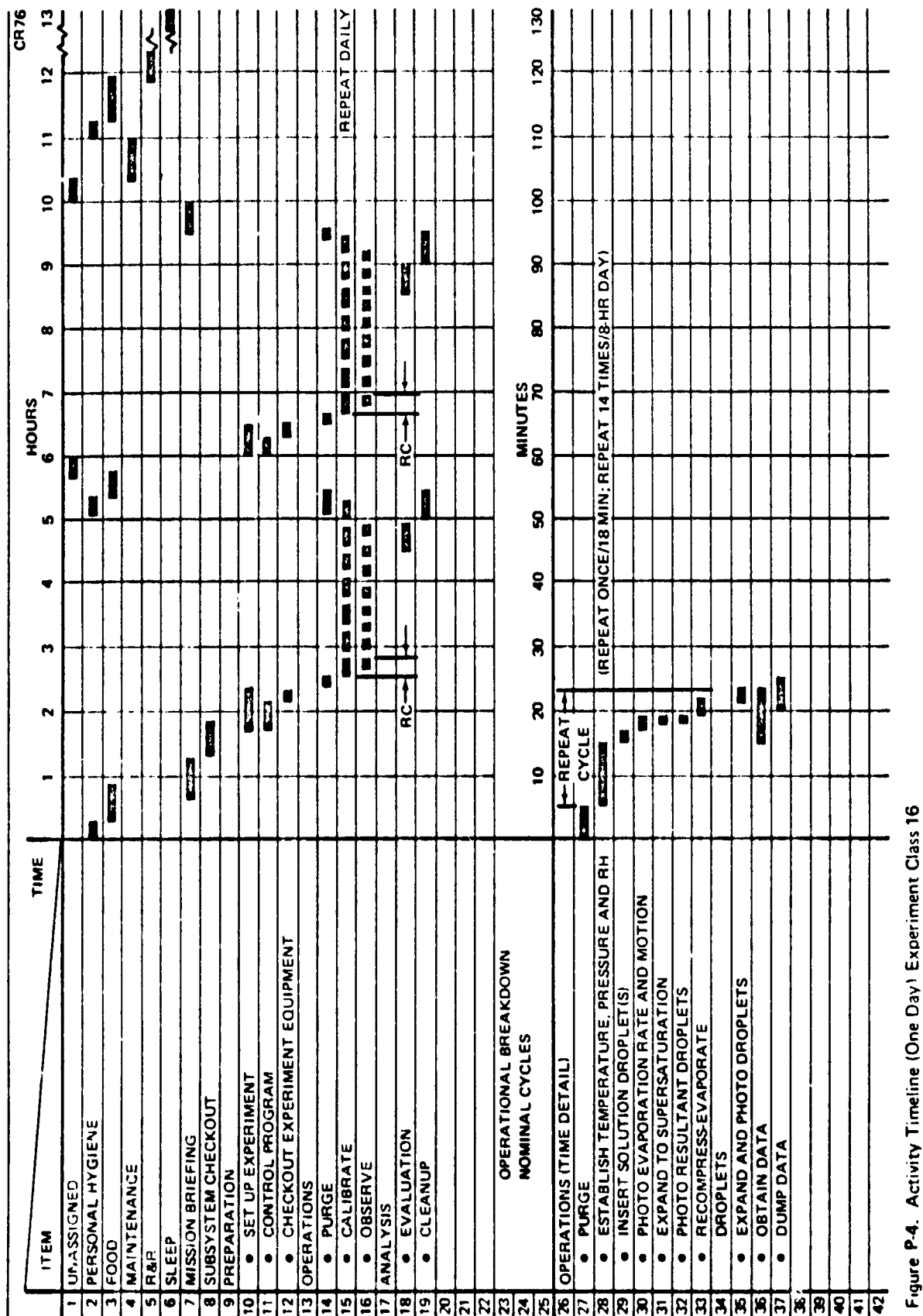


Figure P-4. Activity Timeline (One Day) Experiment Class 16

Appendix Q  
CLASS 17  
DROP COLLISION BREAKUP ( $<0.5 \text{ nm}$ )

Q. 1 DROP COLLISION BREAKUP APPROACH 1

Q. 1. 1 Introduction

There has been persistent, if not continuous, interest in drop impaction research in the cloud physics community. Drop collision experiments have primarily been performed with vertical wind tunnels in recent years. (See reference list). However, MacTaggart-Cowan (1973) has designed a drop electrostatic accelerator that facilitates impactions of drops at variable impact velocities, thus eliminating an experimental dependency on vertical wind tunnels. Edwin X Berry has treated the drop impactions theoretically (to be published, J. Atoms. Sci.).

All these works have helped to quantify impact dynamics in terms of some parameterized variables such as: relative kinetic impaction energies and drop center to drop center separation distance. Quantifying and properly treating the rapid change of aerodynamic drag forces during collision has been the single most glaring deficiency to date of all the experimental and theoretical work done. The influence of aerodynamic forces on coalescence-breakup of drop impactions is monumental. A preliminary demonstration of zero gravity drop collision performed on Skylab has further implicated the role of aerodynamic forces on collision stabilities. Apparently, zero gravity drop impaction experiments are the most promising means of quantifying this effect.

It is well established in cloud physics that warm precipitation is formed through collision and coalescence of water drops. The rate of growth of a droplet with initial radius of  $30\mu$  into a precipitation size particle depends on several parameters: the liquid water content of its immediate environment, the dynamics of the cloud in which it exists, and its present size in the spectrum of other growing drops. It is this third condition which

influences the collection efficiency of the drop and its ability to grow by colliding and coalescing with other liquid drops.

The collection efficiency of drops has been the subject of several investigations. Recently, the studies of impactions between millimeter size drops has had increased interest. The investigations concerning drop impactions have been reported by Magono and Nakamura (1959), Magarvey and Geldart (1962), Gunn (1964), List and Whelpdale (1969), List et al. (1970), Montgomery (1971), Brazier-Smith et al. (1971), and Spengler (1971). All of these experiments introduced some unnatural limitations. Montgomery's and Spengler's work however, were performed on freely suspended drops, thus including important aerodynamic effects. The present studies of drop impactions are designed to eliminate gravitational effects (i. e., aerodynamic forces and hydrostatic pressures) and in doing so, obtain both statistical and specific information concerning the sensitivity of drop impactions to a wide range of variables; drop size, relative kinetic energy, surface tension, electrical effects, point of contact, and viscosity. This knowledge is important for the understanding of precipitation mechanisms. Hopefully it will give some insight into which drop sizes are important to drop collision breakups and coalescence in determining drop size distributions.

#### Q. 1. 2 Objective

The objective of this experiment is to

- (1) Determine and quantify the nature of aerodynamic grab forces on drop impaction results (probabilistic parameters and properties of drop impactions).
- (2) Determine and quantify the influence of other factors on drop impaction outcomes (deterministic surface and fluid properties, external electrostatic or audiosonic fields).

#### Q. 1. 3 Scientific Justification

The understanding of drop impaction is fundamental to cloud physics for both cloud microphysical and thermodynamic processes. Liquid-liquid and liquid-ice collisions have been suggested as charge generating mechanisms. The size distribution of the precipitation spectrum influences the overall

precipitation efficiency of the cloud. Drop impaction apparently plays a dominant role in the growth of drops up to ~2 mm diameter and thereafter begins to retard the growth with non-coalescing impactions. Cleansing processes for removing particles and gases from the atmosphere involves mechanical sweepout, diffusion and/or chemical conversion in the drops.

Thermodynamic properties of clouds involve first order consideration of condensation and latent heat release. However, there has been increasing interest in the effects of evaporation of cloud and precipitation elements and the influence of cloud development. Further, the aerodynamic drag of precipitation elements feeds back as a regulating mechanisms of updraft dynamics.

Because of the importance and implications in cloud physics, pollutant removal and weather modification, a thorough and complete understanding of drop dynamics is needed. Zero-gravity drop impaction studies can provide important information leading to the quantification and relative importance of several potentially influential parameters.

- What is the role of aerodynamic forces on drop collision results?
- How are the energy requirements for bounce, coalescence, and breakup collisions influence by drop surface tension?
- Is charge separation occurring as a result of drop impactions?

These are a few of the specific questions that zero-gravity experiments may provide the answers to.

#### **Q.1.4 Applications**

1. Severe storm cloud dynamics
  - a. Thermodynamic energy considerations as related to drop size distribution.
    - i. evaporation-energy requirements
    - ii. freezing-energy release
  - b. Electrification-charge generation mechanism
  - c. Aerodynamic drag of precipitation elements as a function of drop size
  - d. Icing and hail growth

2. Atmospheric cleansing processes
  - a. Impaction and sweepout efficiencies as a function of drop size distribution
  - b. Chemical transformation of pollutants in drops
3. Inadvertent weather modification
  - a. Change in impaction energy requirements as a function of drop contamination resulting in drop size distribution shifts.
  - b. Change in charge separation rates of colliding drops as a function of contamination
4. Weather modification
  - a. Direct influence of drop size distribution through modification of collision outcomes
    - i. precipitation efficiencies and rate changes
    - ii. thermodynamic changes
    - iii. charging modification
    - iv. hail and icing changes

#### Q.1.5 Terrestrial Laboratory Limitations

In terrestrial laboratories, the need for artificial supports or vertical wind tunnels has always had the limitation in properly simulating some aspects of the natural conditions that may be paramount to the determination of the natural phenomenon. For drop impactions of precipitation size hydrometers, the impact parameters, as discussed by several researchers, are the point of impaction and the (aero-hydro) dynamic stability of the drops. The parameter that remains to be accurately quantified is the influence of aero-hydro dynamic stability of the colliding drop. The rapidly distorting drops, due to collision, are subjected to rapidly changing aerodynamic drag forces. These forces can further distort the drop inhibiting coalescence and hydrodynamic stability.

The influence on colliding drop stability of rapid variations in drag and shear stresses has not been adequately addressed in cloud physics experiments. In a recent article by Sartor et al., the accelerating and decelerating drag coefficient on cloud droplets appears to play an influential role in the coalescence efficiency. This recent experimental interest is an indication of the important questions remaining.

The next iteration of the above mentioned challenge is the quantification of the effects of turbulence. Most wind tunnel experiments have been very careful to specify the characteristic turbulence structure of their air flow. However, no cross comparison has been performed to identify what influence updraft turbulence may have on collision, coalescence efficiencies and drop stability.

#### Q.1.6 Zero-Gravity Opportunities

The zero gravity exploration of drop impaction is quite appropriate in light of the specific information needs discussed above. When considering the limitations of terrestrial investigations, the zero gravity environment affords the best opportunity of differentiating between hydrodynamic (point of contact collision energy, drop distortion) and aerodynamic influences. Once these differences are understood then we can proceed with careful terrestrial experiments designed to quantify microscale turbulence effects.

It is difficult to identify precisely the dollar and time advantages of performing this experiment in a zero gravity environment. It is possible that without zero gravity experiments, some of the basic questions will never be known empirically.

Based on this investigators experimental experience of consuming tens of minutes to hours to achieve at times one data point, zero gravity lab could greatly compress the time requirement for obtaining many collisions. This is important because until the outcome of collisions is determined by quantification of all major influencing parameters, it will have to be studied probabilistically. A great number of precisely recorded collisions would have to be performed. A zero gravity laboratory would facilitate these experiments.

#### Q.1.7 Approach

##### Q.1.7.1 General

The basic problems with terrestrial laboratory studies has been the confounding influence of artificial supports and aerodynamic effects. The



second set of variables relates to experimental uncertainties, drop chemistry and charge, and limitations in the measurement accuracy such as the point of impaction, angular momentum, and mass transfer.

The zero gravity approach to drop collision studies offers improvements in experimental design in both categories of terrestrial uncertainties provided specific experimental and recording equipment is available

#### Basic Equipment Needs

1. Drop generating apparatus with a 100  $\mu\text{m}$  to 20 mm range
2. Drop accelerating device with an upper limit of 20 m/sec
3. Drop stabilization device or chamber to position and hold drops with ease of repeatability
4. Drop oscillator to excite drops into various modes of oscillation
5. Charging device to neutralize or charge the test drops

#### Basic Recording Needs

1. Drop sizing, velocity, and mass measuring equipment or technique
2. Event recording with precision of documenting point of impaction
3. Static charge determination
4. Surface tension recording

#### Q.1.7.2 Detail Requirements

The specific questions being posed in this class of experiments are: what happens when two hydrometers collide and which variables are the most important? To answer these questions within the range of experimental uncertainties a statistical sample of collisions must be performed.

Zero gravity conditions will eliminate aerodynamic effects and the range of experimental variability for several parameters must be conservatively broad so as to ensure all three modes of collision results; bounce, coalescence, and breakup. The experimental requirements include important parameters such as hydrometer size, velocity (which determines relative kinetic energy) as well as the surface tension and initial stability of the hydrometers.

NOTE: Not listed as a parameter per se is the point of impact, i. e., center edge. This variable is very important in determining collision outcomes. However, to build this into the equipment requirements would be extremely difficult, hence, the need for a large number of data iterations to insure collision results for various points of impact. This will require event recorders that can determine the position of impact within a tolerance of approximately  $\pm 0.25$  mm.

#### Q.1.7.3 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	5
Type	
Pollutant	
Pressure	3
Temperature	3
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	5

<u>Parameters</u>	<u>Variations</u>
Liquid-water content	
Surface tension	3
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	5
Gases	
Spin rate	

#### Q.1.8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure Q-1).

1. Set up a general purpose chamber that has predesigned adaptor portals (see Figure Q-2). The chamber is needed more for cleanliness and isolation of drops from laboratory air currents (which was a problem on Skylab). Performing experiments at different temperature and pressure and tertiary variables.
2. Load and ready camera and attach necessary synchronizing cords. The camera should have a variable framing rate and adjustable magnification. The range of framing rates that should be 100 fps to 10,000 fps.

Highspeed requirements stated in NASA CR 129013, (such as framing rates of only 100 fps) appear to be too low for most of the collisions sought in this study. 1000 fps would be more appropriate. A two track system is proposed where one track is used for initial highspeed recording and the other for reviewing the data and storing it. This will allow reuse of highspeed film and storage of just meaningful footage.

3. Positioning the target drop must be an automatic or semiautomatic manually controlled procedure otherwise, tremendous amounts of time will be consumed in performing task.

4. Adjust impacting drop generator for the desired drop size and impact velocity.
5. Set target drop into the desired oscillation mode, if any.
6. Start experiment by releasing impacting drop. This should electronically start the recording equipment with the necessary built in signal delays.
7. Review results of event and decide on storage.
8. Clean up if necessary and run next collision.
9. Shut down apparatus and store.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>● Set up chamber - attach the drop generator-accelerator, with H<sub>2</sub>O reservoir. Attach drop positioner</li> </ul>	5-15
<ul style="list-style-type: none"> <li>● Allow water, chamber to stabilize for T, P</li> </ul>	U
<ul style="list-style-type: none"> <li>● Adjust target drop size, impacting drop sizer and accelerator</li> </ul>	5-10
<ul style="list-style-type: none"> <li>● Load film/video tape on highspeed camera - set up lighting. Attach synchronizer to generator</li> </ul>	3-6
<ul style="list-style-type: none"> <li>● Generate and position drops and apply electrical field if necessary.</li> </ul>	1-10
<ul style="list-style-type: none"> <li>● Generate and accelerate drops while recording.</li> </ul>	seconds
<ul style="list-style-type: none"> <li>● Review event</li> </ul>	3-10
<ul style="list-style-type: none"> <li>● Decide on storing/store</li> </ul>	2-5
<ul style="list-style-type: none"> <li>● Clean up if necessary</li> </ul>	2-4
<ul style="list-style-type: none"> <li>● Repeat</li> </ul>	--
<ul style="list-style-type: none"> <li>● Shut down/cleanup/store</li> </ul>	5-15

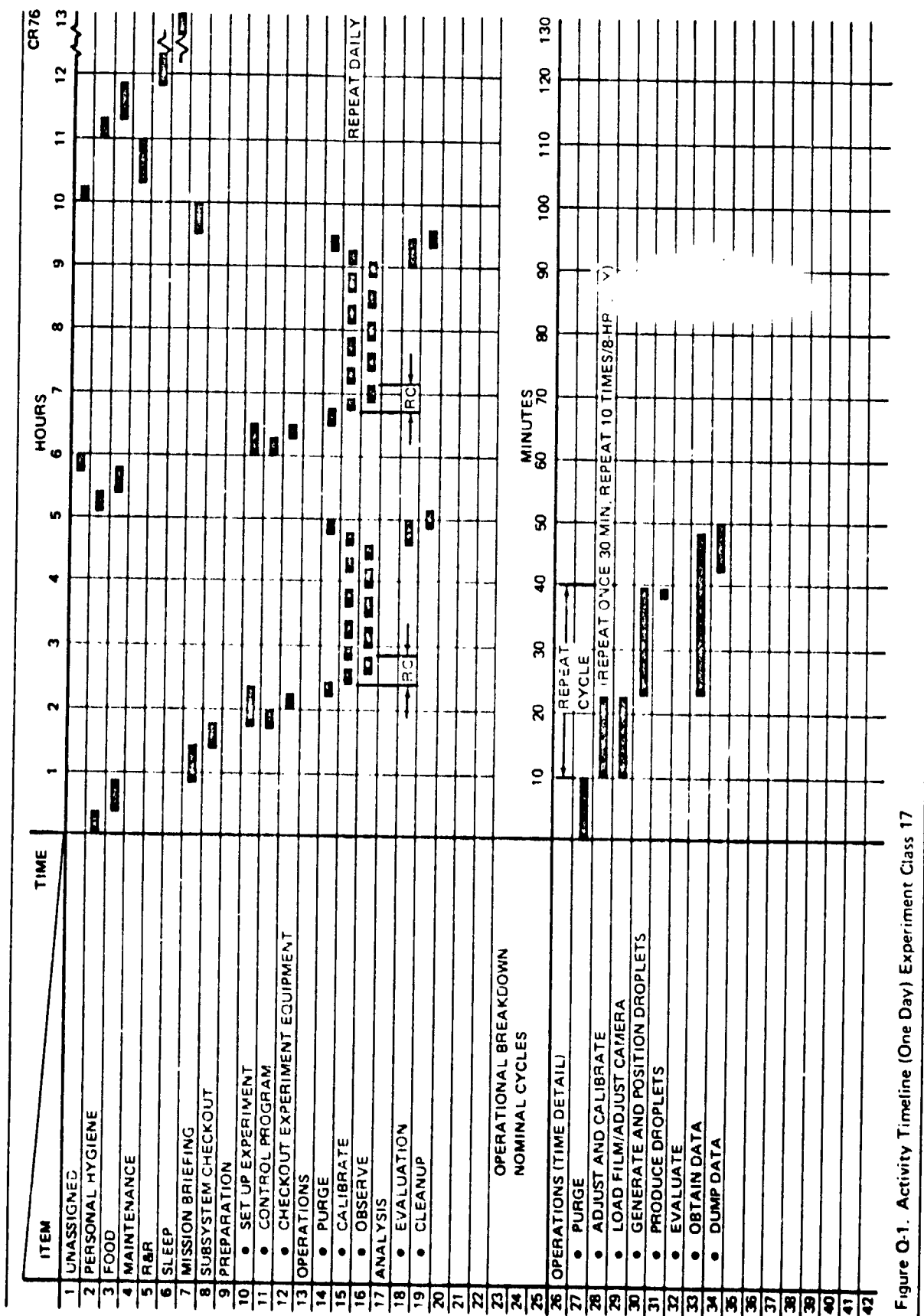


Figure Q-1. Activity Timeline (One Day) Experiment Class 17

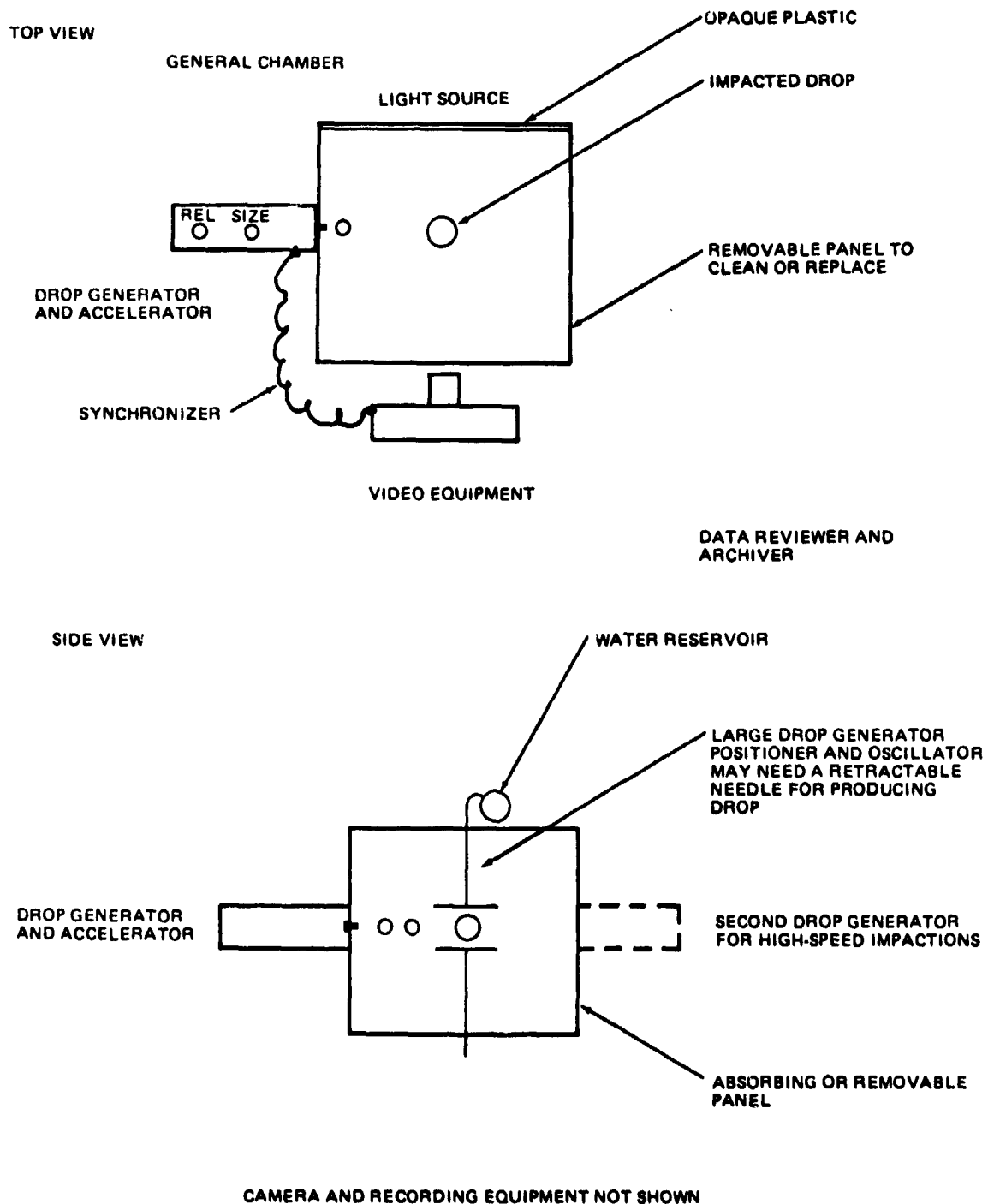


Figure Q-2. General Purpose Chamber

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## Q.2 DROP COLLISION BREAKUP APPROACH 2

### Q.2.1 Introduction

The collision, coalescence and breakup of water drops within a cloud is important in determining the population of large drops present, stability of clouds and of the development of precipitation within them.

The collection efficiencies much greater than unity are found for large drops of approximately equal diameters. When a large drop is falling, the Reynolds number is high and the drop has a pronounced wake in which the air tends to be dragged along behind the drop. The motion of a larger drop is influenced by the wake and it tends to be deflected towards the trajectory of the droplet. This effect is greatest when their sizes are nearly equal, because the deflecting force due to the wake has a longer time in which to act.

It is obvious that if the drops involved in a collision bear unlike charges, the collision efficiencies would be increased while like charges will decrease the chances of collision. Moreover, because water is a dielectric, a strong electric field will induce dipoles upon the droplets which will normally be attractive and hence increase the probability of collision. The question is whether these effects are significant for the charges and fields which may ordinarily exist in clouds.

As suggested by Langmuir, the large drops ( $>5.5\text{mm}$ ) become aerodynamically unstable and breakup into smaller size drops. Another type of breakup, that is well documented and photographed, is due to large droplet-droplet collision induced breakup. It is proposed to study the breakup mechanism under zero-g conditions as a function of fluid properties, drop diameters, electric charges and fields.

### Q.2.2 Objective

The objective of this experiment is to determine the effectiveness and energy requirements of drop-drop collision induced breakup. The different variables in these experiments would be drop sizes, turbulence, electric charges and fields.

#### Q. 2. 3 Scientific Justification

These experiments will provide essential data on energy requirements for drop-drop collision breakup, wake effects and the role of electric charges and fields.

#### Q. 2. 4 Applications

Drop-breakup due to several reasons stated earlier, sets the upper limit to drop-size and determines the distribution. Better understanding of the drop-growth mechanisms within clouds would contribute to our ability to modify precipitation processes.

#### Q. 2. 5 Zero-Gravity Opportunities

The earth's gravitational field restricts detailed observations of the liquid drop breakup processes. The free fall in gravity field is important for these processes, however, to study other variables such as surface tension, viscosity, the air film between the two drops when they make contact, can better be studied under zero-g conditions. Besides, such experiments will provide prolonged observation times and the accurate counting of the number of droplets ejected during collisions.

#### Q. 2. 6 Quantification

It is difficult to predict accurately in advance the progress and success of many scientific experiments. However, as discussed in the earlier sections, the proposed experiments would best be carried out under zero-g conditions.

#### Q. 2. 7 Approach

##### Q. 2. 7. 1 General

A general experiment chamber will be used to obtain these data. Drops of known diameters and kinetic energies will be directed to collide in a low gravity environment. Droplet breakup due to turbulence, drop-drop interactions and electrical forces will be studied with time lapse photography. The ambient variables such as temperature, pressure and relative humidity will be accurately controlled.

#### Q.2.7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	4
Type	3
Pollutant	
Pressure	3
Temperature	4
Relative humidity	4
Charge	4
Rate of cooling	
Time	
Sound	
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	3
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	4
Gases	
Spin rate	

#### Q. 2. 8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlations of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure Q-3).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<p><u>Preparation</u></p> <ul style="list-style-type: none"> <li>● Set up experiment 30</li> <li>● Control program 15</li> <li>● Check out equipment 15</li> </ul> <p><u>Operations</u></p> <ul style="list-style-type: none"> <li>● Purge chamber 5</li> <li>● Establish T, P, R-H 15</li> <li>● Inject and position target drop 5</li> <li>● Start cameras and data recorders 1</li> <li>● Impinge droplet with fixed velocity 5</li> <li>● Recycle with other variables 30               <ul style="list-style-type: none"> <li>- Velocities</li> <li>- Diameters</li> <li>- Trajectories</li> <li>- Surface tensions</li> <li>- Viscosities</li> <li>- Electric charges and fields</li> </ul> </li> </ul> <p>Analysis and cleanup 60</p>	

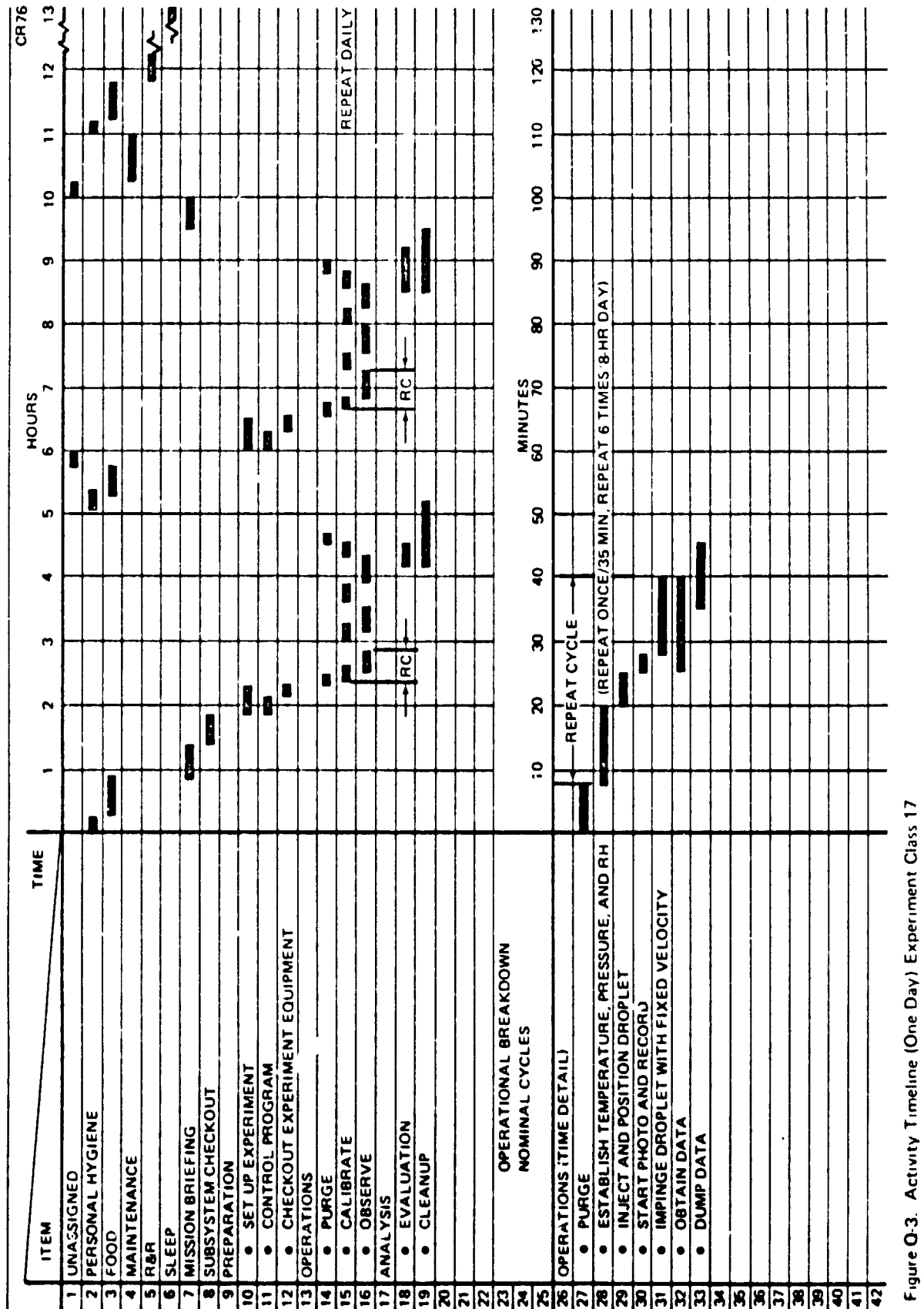


Figure Q-3. Activity Timeline (One Day) Experiment Class 17

## Appendix R

### CLASS 18

#### COALESCENCE EFFICIENCY ( $< 5.0 \mu\text{m}$ )

#### R.1 COALESCENCE EFFICIENCY APPROACH 1

##### R.1.1 Introduction

The overall problem of calculating the rainfall to be expected from a given cloud contains a number of problems yet to be solved, or, even, clearly stated. Single models of clouds which postulated a uniform updraft, showed that a crucial problem in any model was the accurate assessment of collision efficiencies. The problem in its simplest form begins with a distribution of cloud droplets and examines their growth by coalescence resulting from collisions due to the differential fall rates between droplets of slightly different diameter.

The collision probability of two droplets with a given different fall rate and horizontal separation of their centers depends crucially on the dynamics of the air motion during the closest approach. Theoretical solutions are now fairly convincing up to distances of  $1/10$  radius during close approach. However the fundamental question remains as to what distance between the droplet surfaces constitutes contact.

The progress made in theoretical calculations, assuming zero Reynolds number (i. e., inertia in the air plays no role but viscosity controls the airflow), of the interaction between clouddrops (Hocking, 1959 and Davis, 1967), and the extensive experimental studies (Telford, J. W, 1961) have left unanswered questions in this regard. These questions relate to the very last stage just before the separate water surfaces blend. Since the best experiments provide only rather imprecise data because of the small size of the drops and the need to rely on statistical sampling for the measurements, the opportunity to model the collision under low gravity with bigger drops and longer collision times offers the opportunity to greatly improve understanding in this area.



It was evident from the beginning of this work that the experimental difficulties were related to the very small size of the waterdrops and the rapidity with which the collision and coalescence occurred. The small size makes handling of the drops impossible and so good experiments could not be designed involving the interaction of only the two individual drops. Attempts to do this (e. g., Woods and Mason, 1964) failed because the drops were interacting at high speed long before they were photographed at terminal velocity. Thus these results leave serious doubts as to the relevance of the results. The high speed interaction which is present long before collisions in these experiments is not part of the process in real clouds.

At this stage, it was clear that a greatly reduced gravity would be of enormous assistance. The experiment of Telford and Cottis (1969) was designed to simulate very much reduced gravity by suspending the simulated drops by long wires, the tops of which were moved under servo control to keep the wires sloping at a constant small angle. In this experiment, the ratio of the density of the spheres to the fluid had also to be simulated. Although this experiment gave some useful data it was very limited in usefulness, partly because of the crude servo control, but principally because of the drag on the strings supporting the balls in the fluid trough simulating the air.

With the availability of a true low gravity environment we thus have an entirely new field of experiments open to us in those areas where there are still major questions of importance.

One area of particular importance is related to electrical effects between drops freely moving in air. Another relates to the final stages of coalescence of uncharged drops. Since the latter are influenced by electrical effects any experiment will need electrical control equipment and so both types of work probably should be done together.

The fundamental questions to be answered relate to what is happening as the air film between the drops is squeezed out or dissolved and the final forces are those between the polarized surfaces of water molecules. It is well known that drops will skate around a freshly formed water surface for 1/2 second or so. An electric field of  $1 \text{ volt cm.}^{-1}$  or so produces instant

entry of the drop through the flat water surface. Since the weight of the drop itself makes the approach to the other water surface too quick to allow study, an experiment is required using the very much smaller weight force in the low-g environment which will allow time for practical measurements.

It is proposed that two experiments be considered for the zero-g environment.

1. A study of the collision dynamics between fractional millimeter diameter water drops to simulate cloud drop collisions.
2. An experiment with almost flat water surfaces at very close approach (i. e. , 0.025  $\mu$ meters) to examine the instabilities, attraction and coalescence of water surfaces.

#### R. 1. 2 Objective

This experiment is aimed at producing measurements of drop collisions and coalescence under low gravity conditions using larger sized drops which can be scaled to give collection efficiencies at cloud drop sizes.

#### R. 1. 3 Scientific Justification

The modelling of the rain forming mechanism in clouds not involving the ice process is at present largely fitting of speculative mechanisms to our present knowledge of clouds as a whole. One section of these models could be quantified beyond doubt by accurate measurements of collisions between drops representing the cloud drops. This step is essential before a cloud model can be produced which is convincing in the sense that arguments are convincing in other branches of physics. The range of cloud drops of importance is 20  $\mu$ m to 40  $\mu$ m in diameter.

#### R. 1. 4 Applications

The prediction of rainfall from computer models is essential in simulating the atmosphere for the purposes of atmospheric prediction and modification. The results will also have important bearing on pollution scavenging and perhaps even flotation processes which involve collisions between bubbles and other particles.

#### R.1.5 Terrestrial Laboratory Limitations

The fundamental problems in performing this experiment under normal gravity are related to the fact that cloud droplets are too small to be handled or even seen. Indeed the generation of accurately sized  $10\text{ }\mu\text{m}$  diameter drops of uniform size is very difficult and then these drops cannot be touched to position or move them. Convection currents carry them everywhere except where the experimenter needs them and the problem can be seen in perspective when it is realized they need to be positioned to within better than  $1\text{ }\mu\text{m}$  and they fall  $10,000\text{ }\mu\text{m}$  per second. The technique of generating a stream of small drops following a closely defined path defeats the purpose of the experiment since the forces generated by the stream of preceding droplets dominate the dynamics and experiment does not reproduce the motion of two individual droplets starting the interaction at terminal velocities.

#### R.1.6 Zero-Gravity Opportunities

The fundamental difficulties in these type of experiments could probably be solved on earth if gravity could be turned off while the drops were maneuvered into position under a stereo microscope movie-camera arrangement. The air could also then be brought into temperature equilibrium without convection motion. If gravity were then turned on the microscope moved to track the drops we would probably have a practical experiment.

The opportunity to virtually turn off gravity at the center of mass of the capsule would allow this setting up procedure. The turning on of linear gravitational acceleration at  $1\text{ g}$  would be difficult in a capsule because of the large velocities involved. However it turns out that very small controlled accelerations bring major advantages to the experiment in the form of needing larger drops and slower speeds to retain dynamic similarity. The flow appears to be exactly analogous.

Attempts to simulate the collisions using water drops or solid balls of oil fail because although the Reynolds number of the drops may be adjusted to be correct at large separation the Reynolds numbers do not track those parameters of the real drops being simulated when acceleration occurs

during close approach. To be an accurate model the ratio of the drop density to that of the surrounding fluid must be the same as for water drops in air.

For lower gravity the formulae for the terminal velocities of drops are as follows:

Stokes Law gives,

$$\begin{aligned} F &= 6\pi\eta r v = 4\pi(\rho - \rho')r^3 g/3 \\ \therefore V &= 2(\rho - \rho')gr^2/\eta \\ Re &= 4(\rho - \rho')gr^3/g\nu^2\rho' \end{aligned}$$

Where

$$\begin{aligned} \nu &= \eta/\rho' = \text{kinematic viscosity of air} \\ \eta &= \text{dynamic viscosity of air} \\ \rho' &= \text{density of air} \\ \rho &= \text{density of water} \\ r &= \text{drop radius} \\ g &= \text{acceleration due to gravity} \\ Re &= \text{Reynolds number} \end{aligned}$$

Thus, in simulation with reduced  $g$ , using the subscript  $e$  to refer to normal conditions on Earth,

$$\frac{Re}{Re_e} = \frac{gr^3}{g_e r_e^3}$$

Hence, if we plan to set up an experiment at zero  $g$  in a small package and then accelerate the package at  $10^{-3}g_e$  we can simulate using  $200\mu\text{m}$  drops the collision of two really equal drops in the atmosphere, of diameters  $20\mu\text{m}$  (we need to simulate diameters from  $20\mu\text{m}$  to  $40\mu\text{m}$ ).

The terminal velocity of the  $200\mu\text{m}$  drops at  $10^{-3}g_e$  will be  $v = v_e gr^2/g_e r_e^2 = v_e r_e/r = 0.12 \text{ cm sec}^{-1}$ .

Hence the simulated drop moves through its surrounding air at a distance of about 8 radii each second. If we plan on a 1 percent difference in the terminal velocity of two drops, (set up for collision at a distance of 60 diameters apart) they will need a time of

$t = 60 \times 100 \, r/v = 6000 (r_e/v_e)(g_e/g)^{2/3} = 5 (g_e/g)^{2/3}$ , so that at  $10^{-3} g_e$  the experimental drops will take about 10 minutes to collide. If this occurs in a linearly accelerating chamber accelerating at  $g$ , the chamber will move  $L = 1/2 g t^2 = 12 g_e (g_e/g)^{1/3}$  or a distance of 1200 meters or more.

This approach would thus require acceleration of the spacecraft. Sustained acceleration is obtainable from the gravity gradient across the spacecraft. However this is of the order of  $10^{-6} g_e/m$ . This low number suggest an even more desirable experiment where the drops used are  $2000 \mu m$  or  $2 mm$ . in diameter and the experiment runs for a time of 1000 minutes or 16 hours. This would however involve servo control of the spacecraft to keep the experimental chamber in free fall within an experimental volume where the only force on the experimental container is a controlled force generating the  $10^{-6} g_e$  acceleration. Since the  $2 mm$ . drops need to move  $6000 \times 2 mm. = 12 m.$  within their immediate airspace the chamber would also need to be this long and adjusted so the gravitational gradient along it was either negligible or used so the drops always experienced the correct acceleration.

Considerations of trade-offs along this line of thought depend on engineering data unavailable to the author, in particular, the spectral density of the  $g$  noise, which has only been mentioned as of the order of  $10^{-4} g_e$ . Without knowing the excursions in position of the craft around the true central point, nothing further can be done in this way and so an alternative approach is recommended.

Thus the practical approach to using a very low acceleration (e. g.,  $10^{-4} g_e$ ) is to set up the drops in their initial condition and then apply an acceleration by swinging the chamber in a circle.

When the drops have a diameter of  $200 \mu m$  a fall distance of 6000 diameters is 120 cms. and hence the fall space in the chamber would have to be about this length. To achieve an acceleration of  $10^{-3} g_e$  or  $1 cm. sec^{-2}$  the circular path and velocity are given by

$$l = r \Omega^2$$

If  $r = 30 cm.$ ,  $\Omega = 0.18 \text{ radians } sec^{-1}$  or a full circle is  $T = 30 \text{ seconds}$

Thus a free space allowing a circular swing of about 2-1/2 m. diameter is needed for this experiment.

Since such a size may be unwieldy it is suggested that the optimum result will come from choosing the instrument to fit inside a 60 cm. diameter space. Thus the maximum length of fall is about 20 cms with the drops moving on a 10 cm radius circular path. Thus the drop diameters should be  $20/6000 = 33\mu\text{m}$  in diameter.

This size drop calls for an acceleration of  $g_e/4.49 = 218 \text{ cms/sec}^2$ .

If the radius arm is 10 cms  
=  $(218/10)^{1/2} = 4.67 \text{ radians/sec.}$   
T = 1.35 seconds per revolution.

The time needed for the drops to fall is  $t = 14 \text{ seconds.}$

Thus while the drops are smaller than could be desired the experiment looks practical to perform under low gravity conditions. At these substantial accelerations the movement of the spacecraft would be negligible.

Let us repeat the exercise with half the initial separation of the drops which may be sufficient in most cases. If 20 cm. is 3000 drop diameters the drop will be  $67\mu\text{m}$ . Thus we need an acceleration  $g_e/38 = 26 \text{ cm. sec}^{-2}$ .

Thus for a 10 cm. radial distance of the rotating chamber  
= 1.6 rad/sec  
T = 3.9 secs.

The time for the drop falling is  
t = 44 seconds

This final configuration looks the best particularly if a total diameter greater than 60 cms is feasible.

In the proposed experiment, using optical observation, the position of two drops will be adjusted using small air jets or the centrifugal acceleration gradient while on the axis of rotation of a transparent square tube rotating around a line perpendicular to the tube through one end. In this essentially non-accelerated region the task of adjusting position can be done at leisure and may well take 10 to 15 minutes to set up the required drop trial. This overcomes the problem met in terrestrial experiments associated with the impossibility of positioning the participating drops as needed.

Acceleration is then applied to the drops by moving the transparent square tube along its axis away from the axis of rotation until the drops themselves are 10 cms. from the axis of rotation. Here the acceleration is  $26 \text{ cm/sec}^{-1}$  and this is maintained by moving the transparent tube back along its axis so the drops are always 10 cm. from the axis of rotation.

Since we are considering  $67\mu\text{m}$  diameter drops 30 diameters apart this is a separation of 2 mm. in 10 cms. and the centrifugal acceleration difference is 2 percent which is negligible. Similarly the gas will have virtually no motion induced by these accelerations.  $g_e$  gives at 1000 mb pressure a pressure gradient of  $0.12 \text{ mb. m}^{-1}$ . The effects in this experiment on gas compression are thus very much less than 1 in  $10^4$  and so are negligible. Gas temperature and moisture differences should be evened out by stirring between each experimental run.

The drop motion can be photographed with two perpendicular views superimposed in color on the film.

It is essential for the success of this experiment to provide the operator with a good viewing and manipulation facility. The essential capability this experiment offers is the ability to initialize the drop motion precisely as needed without throwing a stream of drops into place along a track, or picking pairs at random or using some other expedient which has so seriously hampered terrestrial approaches to this measurement problem.

#### R. 1.7 Justification

Atmospheric science is a young science and thus is filled by speculation, advocated assertions and claims that problems have been solved supported only by the redefinition of the standards of success. This type of situation has occurred in science before, as in the study of chemical equivalents in the Nineteenth Century, for example, when the situation was sorted out only by precise measurements. It is important for the atmospheric sciences to mature to a quantitative science and this experiment provides an opportunity to move towards quantifying a crucial step in rainmaking theory. This experiment is beyond foreseeable technology to perform on earth and will probably be left aside if not done in orbit.

This experiment can be used to evaluate electrical effects and the effects of surface active agents on the coalescence process. These are possible avenues for control of the process of rain formation.

#### R. 1.8 Approach

##### R. 1.8.1 General

This experiment is concerned with simulating the dynamics of cloud drop collisions and requires a special apparatus and may be situated anywhere there is power available. The experiment follows the work of Telford from 1950 to about 1957 when it was given up because of technology barriers. Later work involves theoretical calculations (or non analogous models used to support theories which are then used in the real drop calculations).

While the experiment basically involves uncharged and uncontaminated drops, provision should be made for charging electric fields and surface contaminants (e. g., dissolve hexadecyl trimethyl ammonium bromide, ethyl alcohol, etc. in the water used at different stages).



## R. 1. 8. 2 Experimental Apparatus

### The Drop Generator

Use a Vibrating Needle with some method of allowing one uncharged drop at a time into the chamber (e. g., perturb vibration mechanically or magnetically.

A simple procedure for charging drop sizes must also be built in.

### Stirring

A stirring paddle could permanently remain in the enclosure and be moved to stir. A lid could be opened at each end of the enclosure so with an enclosed rotating frame the radial movement of the enclosure would stir the air in it. Provision also needs to be made for wiping the inside walls clear of condensation.

### Temperature

Substantial heating by the lamps must be prevented by efficient filtering and cooling of the lamp itself.

### Digital Display

A small digital display should superimpose day, time, measured angular rate, rate of radial movement of enclosure, and sideways displacement of enclosure at axis. Also simulated drop size and other variables may be desirable. It would be preferable to have a green display so the drop images could be red or blue. However a red display with green and blue drops is probably satisfactory.

### Movie

Recording should be done by Color lapse time photography. Frame rates of 2 or 3 per second should be satisfactory.

### Motion Control's

A constant but selectable angular rotation rate should be provided. If we have a 1:8 range in angular rates we can simulate 20 $\mu$ m to 40 $\mu$ m cloud drop diameters with drops about 67 $\mu$ m in diameter.

The rotation periods will thus range from 4 secs to 1/2 sec.

The experimental fall times will range from 44 secs to 4-1/2 secs.

The motion of the enclosure will require simply operated remote controls. The motion must not be jerky or show vibration. Small motion of radius up to 20 mm. under joystick control is needed when on the axis during "setting up". This will allow some movement of the drops using centrifugal force.

Set up will also require a gentle air jet in 3 directions to position one of the drops relative to the other.

When the drops are set, a smooth action will be necessary to move the enclosure 10 cm. out along the rotating radius and commence tracking at the drop's terminal velocity. The tracking velocity will then need to be adjusted manually to keep the lower drop exactly at 10 cm. This motion needs to be predictable as corrections may be needed in the initial positioning to take account of the coriolis force on the drop when moving out to 10 cm.

Projected graticules will be needed to give the drops position in the enclosure and the center of rotation and the exact direction of the radial acceleration.

#### R. 1. 8. 3 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-Nuclei	
Size-Droplet	3
Type	3
Pollutant	
Pressure	
Temperature	
Relative humidity	
Charge	
Rate of cooling	

<u>Parameters</u>	<u>Variations</u>
Time	
Sound	
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	6
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### **R.1.9 Procedure**

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure R-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<u>Falling Drops</u> <ul style="list-style-type: none"> <li>● Set up and start generating drops; clean and saturate enclosure, load camera, etc.</li> <li>● Stir and wipe enclosure</li> <li>● Select and position drop pair</li> <li>● Follow drop fall</li> <li>● Change solutions</li> <li>● Hook up and test electric fields</li> </ul>	<div>20</div> <div>1</div> <div>10</div> <div>1</div> <div>20</div> <div>20</div>

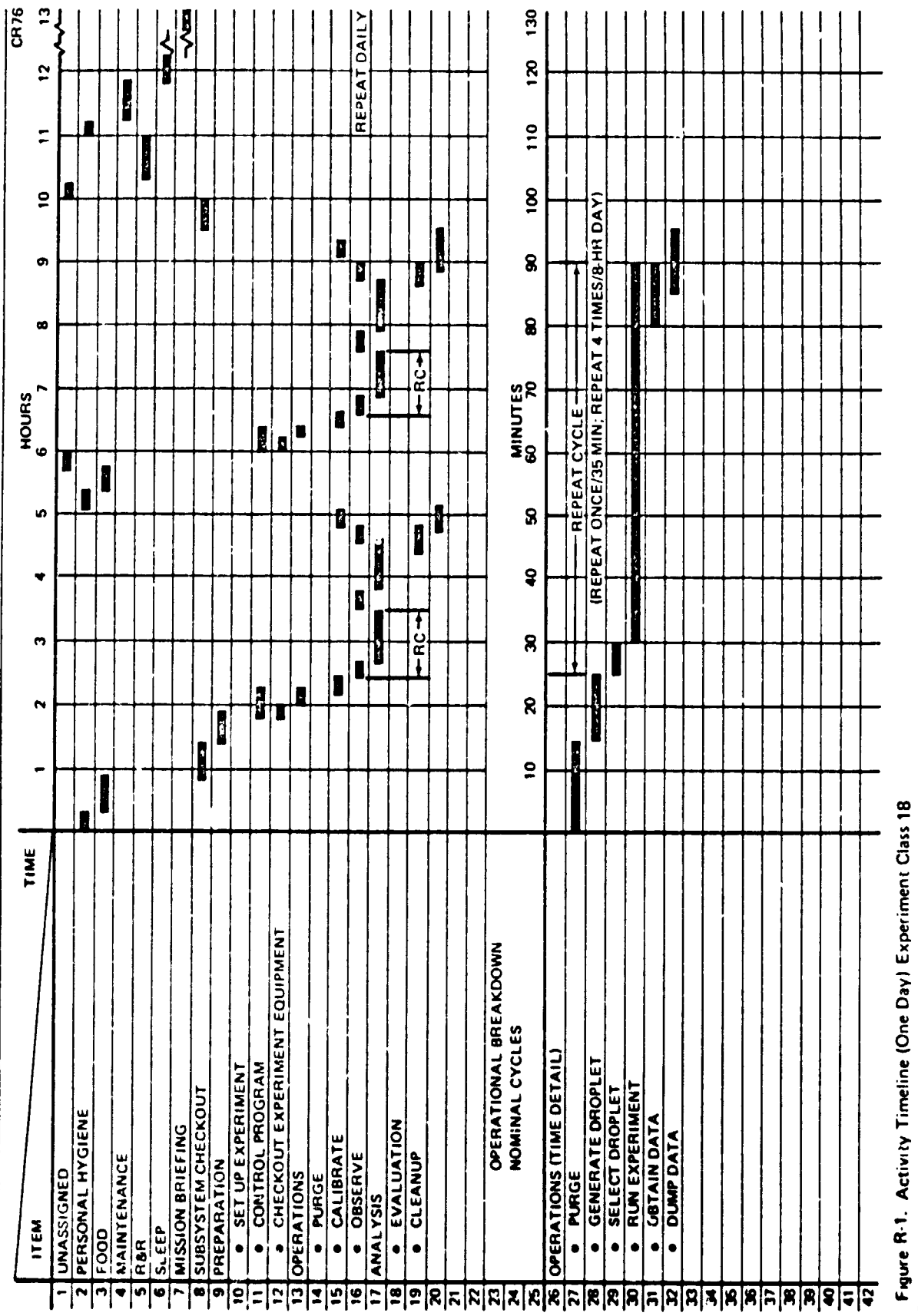


Figure R-1. Activity Timeline (One Day) Experiment Class 18

## R.2 COALESCENCE EFFICIENCY APPROACH 2

### R.2.1 Objective

This experiment is aimed at casting light on an essential part of the collection efficiency problem, for example, how the actual combination of the two water drops commences.

### R.2.2 Scientific Justification

The least appreciated section of the problem of cloud droplet coalescence, because so little is known about it, is the actual process where the two surfaces of the approaching drops blend together. The proper modeling of the airflow produces infinite repulsive forces as the gap tends to zero. Indeed the dynamic gas bearing uses similar forces to support substantial loads. This suggests most contacts between solids are at high spot irregularities. For water surfaces the question is whether the initial bridge occurs due to an instability of the liquid surface bridging the gap in small spots, due to the vapor diffusing into the gap so that embryo water surfaces form in the gap or the gas dissolving in the water; or does the coalescence depend on micro-particles on the water surface? Measurement using Newton's rings, the bridging of the gap by a totally reflected internal light wave at about a spacing of  $\lambda/20$ , and an average capacitive gap measurement are proposed. The two optical methods both image the whole surface.

It is conceivable that an understanding of this process will lead to ideas for inhibiting or enhancing it. This could lead to the control of the rainforming process.

### R.2.3 Applications

The control of the coalescence mechanism of formation and the basis of one aspect of an adequate theory of rain formation.

### R.2.4 Terrestrial Laboratory Limitations

The author has examined, over the past 20 years, a number of possible ways of looking at this problem without any prospect of success. The difficulties are that small drops are stable enough because of surface tension but are quite hopeless from the point of view of evaporation and getting an optical

system inside them to see what happens at the point of contact. Larger drops skate on a water surface but their path is quite unpredictable and once again cannot be optically examined because the event of merging does not occur where the instrument can see (with any reasonable probability). In addition the event is so quick no progress of the merging could be seen without high speed photography and literally miles of film would be needed if any hope of seeing an event were to be likely.

#### R.2.5 Zero-Gravity Opportunities

The solution to the observational problems can be obtained if a water surface about 5 mm. in diameter can be gradually brought up to another flat water surface of similar diameter. Under earth's gravity a lower, level surface is quite stable but there is no way the overlying surface can remain fluid and be stable and easy to control.

#### R.2.6 Quantification

In this experiment there is no conceivable chance on earth of bringing two free parallel surfaces of water together to separations of  $10^{-7}$  m. or so. This seems an obvious way to study surface forces and the structure of the water surface but it is not likely to advance in terrestrial laboratories before the development of much more sophisticated devices to use hanging drops of exactly the right size to allow focussing on the interface and very fast recording techniques to examine the small short-lived area of interest. Hence the experiment will probably not be attempted, since the information about surface instability would be tied up in the dwell time before contact which would be hopelessly tied up with vibration and the mechanical movement involved in bringing the surfaces together.

In a zero g field, the surfaces can be brought together within a few fringes and from here on the interposing gas layer should help stabilize the gap if the space is closed up very slowly. Vibration will be a problem if present and care will be needed to avoid this. Vibration isolation can achieve the ultimate in free fall where no dead weight need be suspended.

### R.2.7 Approach

#### R.2.7.1 General

This experiment is designed to examine flat water surfaces in close proximity. The exact conditions of temperature and humidity are not important although temperature gradients need to be avoided.

The apparatus provides a means of bringing two water surfaces together and adjusting the spacing between them by observing Newton's rings and making minute adjustments to the surface curvature by volume changes. A precise method of changing the water volume through the canals, which is not subject to temperature effects (i. e., expansion, thermometer-like, effects) is needed.

Thus any instabilities of the order of  $1/2$  wavelength of light will clearly be seen.

At closer spacings the final breakthrough will be made visible by using a bright neon laser light with internal reflection (the Newton ring is black close up). When the gap is less than about  $\lambda/20$  the surface light wave generated by the internal reflection leaks through and the surface becomes increasingly transparent at places where the spacing becomes less than this value. This procedure should show the final spots where coalescence starts.

High frequency capacitive measurements should be provided to allow average values of air gap to be measured although this may introduce instability from the electrical forces.

The experiment should be performed with temperature differences between the surfaces and surface active agents in the water (e. g., hexadecyl trimethyl ammonium bromide).

Since Fletcher's work has suggested peculiarities in the surface layer of ice (see also Telford and Turner) it would be interesting to do the experiment with one and both surfaces frozen.



Experiments with other fluids may be worth while.

Microsecond voltage pulsing across the surface should be provided since this may show up dynamical changes at the surface whereas a continuous voltage would produce such instability that the surfaces could not be brought close together.

Digital data giving time, temperature, solution number, etc. needs to be superimposed on the color film using a green digital LED display.

#### R.2.7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	
Type	3
Pollutant	
Pressure	
Temperature	15
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	3
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	

<u>Parameters</u>	<u>Variations</u>
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### R.2.8 Procedure

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure R-2).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<p><u>Water Surfaces Near Contact</u></p> <ul style="list-style-type: none"> <li>● Set up, wet surfaces, adjust lights, load camera, etc.</li> <li>● Move surfaces to get Newton's rings</li> <li>● Adjust water for parallel surfaces) Adjust prism for parallel surfaces)</li> <li>● Slowly move surfaces together (0.01 <math>\mu\text{m}</math> steps) Watch for contact</li> <li>● Change solution</li> <li>● Hook up and test for pulsed Electric Fields</li> </ul>	<p>20</p> <p>10</p> <p>10</p> <p>10</p> <p>20</p> <p>20</p>

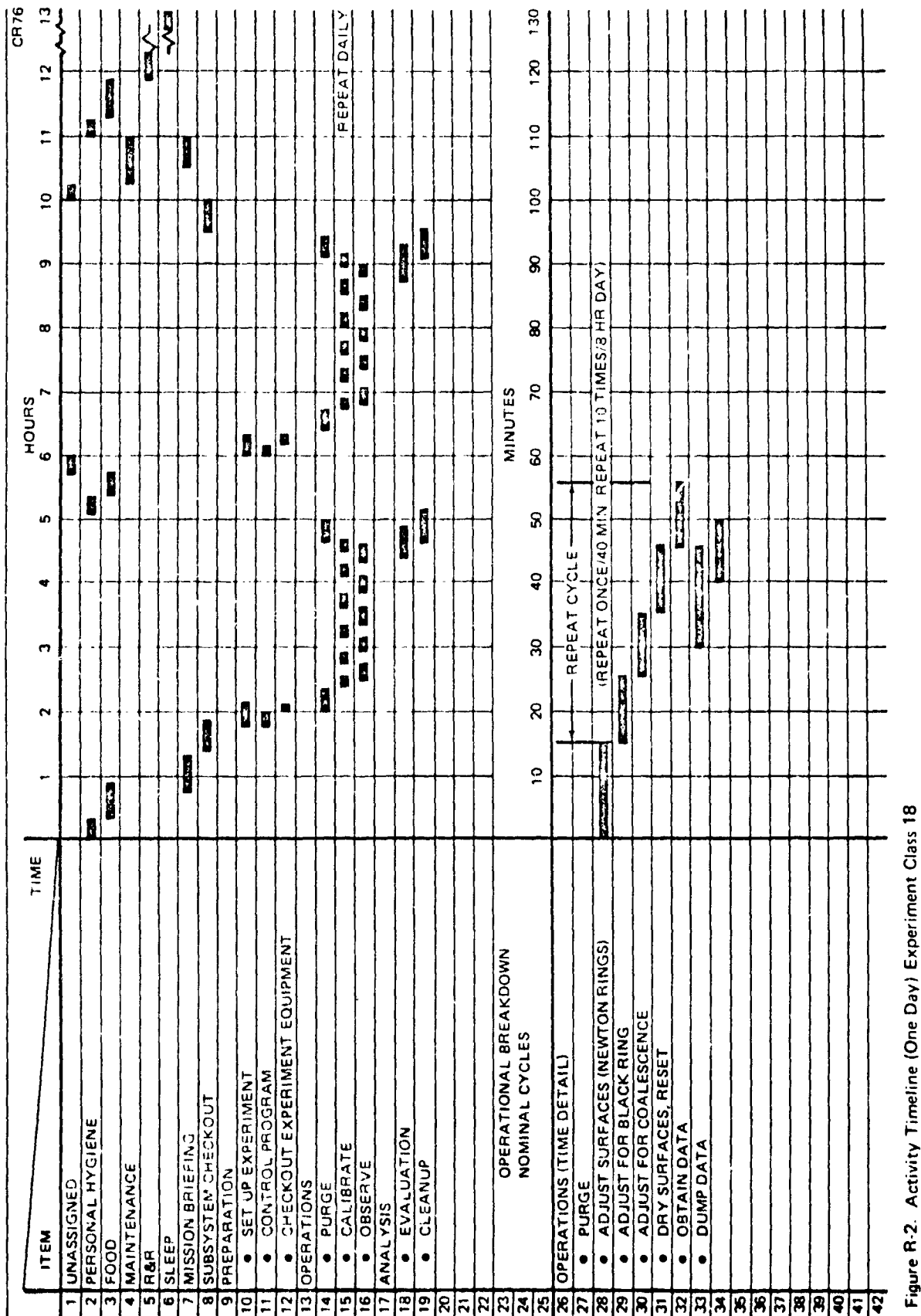


Figure R-2. Activity Timeline (One Day) Experiment Class 18

### R.2.9 References

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## R.3 COALESCENCE EFFICIENCY APPROACH 3

### R.3.1 Introduction

The stochastic coalescence approach has been used during the past decade to mathematically model precipitation formation in warm clouds where the dominant process is assumed to be the development of precipitation size particles through the mutual coalescence of cloud droplets. It now appears that a somewhat similar process may be responsible for the development of the "graupel" particles around which hail stones are able to grow. The stochastic coalescence problem has been well analyzed by a number of writers during this decade, particularly by Mason (1971)<sup>1</sup>, Hidy and Brock (1970)<sup>2</sup> and Gillespie (1972)<sup>3</sup>.

In the standard stochastic coalescence treatments only hydrodynamic-gravitational forces are considered to be primary, however, in the experiment the turbulence is included. Mathematically, it has been shown, that provided the levels of turbulence are significant, considerable particle interactions occur presumably leading to coalescences in size ranges (10 $\mu$ m - 30 $\mu$ m) where standard stochastic theory predicts little or no interactions.

### R.3.2 Objective

The objective of this experiment is to test experimentally the theoretical conclusions regarding the influence of known levels of air turbulence on the

stochastic coalescence between two populations of droplets each having a mono-dispersed size distribution in the size range  $10\mu\text{m}$  and  $30\mu\text{m}$ . By carrying out the experiment in a low gravity environment the contribution to the coalescence by gravity will be minimal and thus any observed coalescence must be attributed to turbulence induced interactions.

### R.3.3 Scientific Justification

The experiment will be of great significance to the scientific community because it will demonstrate that the present so-called "gap" in the cloud particle growth spectrum may be overcome by hydrodynamical-gravitational-turbulence coalescence. Provided this is shown to be experimentally demonstrable, the basic concepts regarding rain formation in maritime tropical clouds and hail formation in continental clouds will have to be revised to take into account the influence exerted by air turbulence. It is conceivable that future rain inducement and hail suppression experiments may utilize this new understanding by designing operational seeding methodologies which seek to exploit the turbulence factor.

### R.3.4 Applications

1. Stochastic coalescence theory for particle pairs in the size range  $10\mu\text{m}$  to  $30\mu\text{m}$
2. Formation of rain in warm clouds
3. Formation of hail stone cores
4. New methodology for rainfall inducement in warm clouds
5. New methodology for hail suppression
6. New methodology for clearing fogs

### R.3.5 Terrestrial Laboratory Limitations

Theory predicts that the effects of turbulence induced coalescence will be evident in about 20 minutes if the turbulence has an energy dissipation factor,  $\epsilon$ , between 1 and 10. Obviously this amount of time required before these effects can be seen would require a terrestrial cloud physics chamber of considerable height due to the fall distance under normal gravitational forces. The following table illustrates the dependence of suspension time

on the gravitational force. In this computation it was assumed a water droplet of 30 $\mu$ m radius

gravity value	g normal	$10^{-2}g$	$10^{-3}g$	$10^{-4}g$	$10^{-5}g$
observation time	1 second	90 sec	1000 sec	$10^4$ sec	28 hrs

It is evident that the time domain requirement for our experiment is best met by a gravity environment of  $10^{-5}g$  or smaller. In addition to the time domain aspect, we wish also to clearly separate the normal hydrodynamic-gravitational stochastic coalescence from the proposed hydrodynamic-turbulence stochastic coalescence. The only feasible way to clearly isolate the latter mechanism is in an environment where gravity forces are minor (less than  $10^{-3}g$ ).

If a turbulence coalescence experiment were performed in a terrestrial laboratory (i. e., in a very tall cloud chamber) the results would be open to criticism because of the ambiguity introduced by gravitationally induced coalescence. Certainly no clear conclusions would be drawn, and moreover the chances are great that erroneous conclusions may be reached as to the role of turbulence induced stochastic coalescence.

#### R.3.6 Zero-Gravity Opportunities

The proposed zero-g cloud physics laboratory would provide the kind of environment needed to test the hydrodynamic turbulence stochastic coalescence theory. It offers a gravity environment ranging from  $10^{-2}$  to  $10^{-5}g$  which covers the value of  $10^{-3}g$  to  $10^{-4}g$  needed for our experiment observation time. In addition, the low gravity field will clearly identify any observed coalescence as being due exclusively to the influence of turbulence.

#### R.3.7 Quantification

The proposed experiment has no parallel in an earth bound laboratory, therefore, it is not possible to reach similar conclusions simply by extending the time frame for the terrestrial analogue.

### R. 3. 8 Approach

#### R. 3. 8. 1 General

Work in the stochastic coalescence problem has been restricted to the hydrodynamic-gravitational formulation which according to a recent review by Mason<sup>1</sup>, accounts for the growth of cloud droplets through the collision of pairs of droplets. The existing theory shows that little or no collection occurs when the pair partners are in the size range  $10\mu\text{m} - 30\mu\text{m}$  radius. The proposed theory which adds the influence of turbulence on the particles' trajectories, shows significant collision probabilities in this size range. The problem is to obtain experimental evidence which confirms the theoretical findings.

Several approaches could be used but the probable success is highly doubtful in all but the gravity controlled experiment. The basic problems are (1) to control the initial sizes of the cloud particles so that they are in the range  $10\mu\text{m} - 30\mu\text{m}$  radius, (2) to create a level of turbulence within the cloud having a known energy dissipation factor, (3) to monitor the subsequent particle size evaluation over a time interval long enough for the turbulence influence to manifest itself, and (4) to be able to separate the influence of gravity induced collisions from turbulence induced collisions. If the experiment were performed in a terrestrial laboratory, the observational time required would be most difficult to achieve due to the settling of the particles due to the gravitational field. In addition, considerable difficulty is anticipated in trying to separate out the small but not insignificant collision probability due to the gravity field itself.

Therefore, the ideal experimental is the one offered by the zero-g cloud chamber. Under conditions of  $10^{-3}g$ , droplet collisions could be monitored for periods up to 1/2 hour without appreciable settling. Using the general purpose chamber with acoustic drivers, turbulent air fluctuations could be induced to simulate cloud turbulence levels appropriate to the theory. Light scattering or direct photographic techniques can be used to monitor the evolution of the drop size distribution in the chamber. Ancillary equipment proposed for the chamber would provide the droplets of specified initial sizes and number. The entire experiment could be electronically programmed



and recorded so that an astronaut need only to push buttons or turn dials to start up or stop the experiment. Subsequent analysis of the results would be performed from the magnetic tapes.

#### R. 3. 8. 2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	2
Type	
Pollutant	
Pressure	3
Temperature	2
Relative humidity	4
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	5
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

### R.3.9 References

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Appendix S  
CLASS 19  
STATIC DIFFUSION CHAMBER EVALUATION

S. 1 INTRODUCTION

In recent years the thermal diffusion cloud chamber patterned after that of Langsdorf (1936), Wieland (1956) and later Twomey (1963), has found a wide range of uses by cloud physicists and atmospheric scientists. Cloud nucleus chambers have contributed to our understanding of the number and type of condensation nuclei that are active in clouds and fog and in identifying certain pollutants that contribute to inadvertent weather modification. Since Twomey's measurements of cloud nuclei (CCN) in the late fifties using a chemical gradient diffusion chamber (1959), steady improvements have been made in the design and operating features of CCN counters. More recently automated, as well as continuous flow diffusion chambers, have been developed. Although there have been a number of attempts to use the simpler expansion chamber for CCN measurements (i. e., Gardner Assoc. Small Particle Detector), the static diffusion chamber (SDC) continues to be the standard against which other CCN measurements are compared.

At the 2nd International Workshop on Condensation and Ice Nuclei (IWICN) held in Ft. Collins, Colorado in August 1972, an unusual opportunity arose for comparing chambers of similar design. A total of five diffusion chambers were brought to the workshop by various workers in the field. Using a wide range of particle sizes and types, comparisons were made of the observed concentration of cloud nuclei at supersaturations (S) ranging from 0.3 percent to 1.0 percent S (i. e., RH 100.3 percent to 101 percent). In general, the measurements agreed to within  $\pm 25$  percent, substantially better than the factor of 2 observed for Aitken counters and the several orders of magnitude difference observed among the ice nucleus counters.

In spite of the good agreement over the range of supersaturations tested, no attempts were made to operate the SDC at supersaturations  $< 0.2$  percent, a value which is thought to be representative of the relative humidities found in most stratus clouds and fog. Regrettably, the operating range of the SDC is limited to about 0.2 percent S in the terrestrial laboratory, for at lower supersaturations the growing droplets fall out of the sensitive volume before they can be detected by photographs or other image sensing devices.

The Zero-G environment provides the experimenter with a unique opportunity to observe droplet growth over extended lengths of time and to measure nucleating efficiencies of particles at saturations less than 0.2 percent. Comparisons using standardized nuclei sources in Zero-G and 1-G environments may allow correction factors to be derived which can be applied to the terrestrial SDC in order to extend its operating range to lower supersaturations. The comparative evaluation will also help identify the measurement accuracy of the SDC when observing nuclei of mixed chemical composition and, in particular, when observing slow to activate hydrophobic nuclei.

## S.2 OBJECTIVE

The main objective of the proposed evaluation is to determine the absolute nucleation efficiencies of standardized nuclei sources under zero fallout conditions using a static diffusion chamber. A secondary objective is to provide correction factors that will help extend the useful operating range of the terrestrial cloud nucleus chamber.

## S.3 SCIENTIFIC JUSTIFICATION

Most theories of cloud droplet formation require knowledge of the critical supersaturation of a nucleus, for it is this property which determines if a particle, once activated, will grow without limit to larger sizes. A thermal diffusion chamber provides information on the nucleation efficiencies of particles at supersaturations that are comparable to those found in most clouds. Past studies have generated useful information on the cloud nucleus behavior of natural as well as artificial (i. e., seeding) aerosols. This information has helped the experimenter and dynamicist to interpret cloud formation and precipitation processes and to assess the effects of seeding on fog and cloud modification.

Unfortunately, the researcher working on fog modification problems or on fog life cycle studies has been hampered by an inability to make measurements of cloud nuclei which correspond to the observed natural droplet concentrations. Usually this is because the SDC cannot be operated at sufficiently low supersaturations. The zero-G atmosphere provides a fallout free environment that can be used to determine absolute nucleation efficiencies of standardized nuclei sources for comparison and calibration with terrestrial chambers.

#### S.4 APPLICATIONS

- (1) Use the data to evaluate the absolute accuracy of the terrestrial SDC and provide knowledge on the early droplet growth behavior of natural and artificial nuclei at very low supersaturation (e. g. , 0.05 percent).
- (2) Utilize the data to interpret the nucleation efficiencies of common seeding agents (i. e. , AgI, NaCl) used in weather modification.
- (3) Help provide an interpretation of the effects of atmospheric pollution on nucleating efficiency.

#### S.5 TERRESTRIAL LABORATORY LIMITATIONS

The operating range of the static diffusion chamber is limited in the terrestrial laboratory because of droplet fallout from the sensitive volume. Increasing the separation distance between the warmer upper surface and the cooler lower plate does not help since an even longer time is required for the specific supersaturation state to be reached (even if the aspect ratio; i. e. , diameter to height, is kept above 6). Thus in the terrestrial environment, a lower limit is imposed on the operating range of the SDC which prevents the acquisition of important data on nucleation efficiencies of particles in clouds.

#### S.6 ZERO-GRAVITY OPPORTUNITIES

In light of the foregoing discussion, it is obvious that the fallout free zero gravity environment provides a very special opportunity for observing droplet growth in the SDC for extended periods of time and for determining the nucleation capabilities of particles at lower supersaturations. Although these data

have not been obtained in the terrestrial laboratory, it is possible that with the help of the zero-G experiment, interpretation of future data acquired at low supersaturations will be vastly improved. For example, in previous studies involving fog modification we have not been able to make comparative measurements between number of nuclei activated and droplet concentrations in fog. Similarly, observations of the interaction of pollutant aerosols with clouds and fog have been limited by an inability to acquire data over a broad enough spectrum of supersaturations. The zero-G environment provides the framework for a comprehensive evaluation of the SDC and also a chance to derive absolute comparisons of chamber efficiency. This information can be obtained by using equipment that exists today and can be used to extend the overall operating range and utility of the SDC in the terrestrial laboratory. Several years of costly equipment development time might be saved.

## S.7 APPROACH

### S.7.1 General

The basic approach will be to obtain data on the nucleation efficiencies of standardized nuclei sources in the zero-G environment for comparison with data obtained using identical procedures and nuclei in the terrestrial laboratory. Comparisons of these results will help quantify the errors due to operating static diffusion chambers in 1-G environments.

Previous work directed toward the evaluation of the static diffusion chambers has been outlined in the Introduction. From an experimental point of view only the 2nd International Workshop on Condensation and Ice Nuclei has provided an adequate opportunity for interchamber comparisons. Very useful theoretical treatments of diffusion chamber design and operation have been provided by Twomey (1959, 1963), Saxena (1970), Fitzgerald (1970) and Squires (1971). While the previous studies have been instrumental in our understanding and proper use of the SDC, there still has not been an opportunity to make a comprehensive evaluation of the static diffusion chamber. The zero-G environment provides this opportunity.

Data will be obtained with a SDC employing photographic recording of droplets. Prior to zero-G experiments, the instrument will be calibrated against other CCN counters using standard aerosols. Improved methods of particle production must still be derived; however, an acceptable procedure for soluble nuclei can be built around the use of a monodisperse aerosol generator or vibrating orifice. Nuclei of the desired sizes can be produced by using carefully prepared solutions of a soluble salt, such as NaCl, and relying on evaporation of the generated solution droplets in a conditioning chamber or other holding vessel. Good reliability can be achieved in obtaining nuclei in the size range of 0.005 to 0.1  $\mu\text{m}$ .

Samples containing suitable numbers of nuclei may be introduced into the SDC either directly or from the conditioning chamber. Relatively low concentrations of particles should be produced in order to minimize coagulation during the short storage time in the conditioning chamber. Several discrete supersaturations will be tested both in zero-G and in the terrestrial laboratory. Particle size distribution analysis will be provided by an Electric Aerosol Analyzer manufactured by Thermo-Systems, Inc., Minneapolis, Minnesota.

For the most part, direct comparisons of nucleation efficiency can be made between instruments in the range of supersaturations from 0.2 percent and 2.0 percent S. Below 0.2 percent S, only the zero-G operated chamber is expected to provide reliable data. This information will be helpful for correlations with observed nucleus concentration in the terrestrial laboratory. Special emphasis during these tests will be placed on obtaining repeatability of test results in order to insure statistically valid comparisons.

An alternate approach to using a single SDC would be to employ one SDC and one CFD chamber. The two instruments may be calibrated side by side in the terrestrial laboratory and then compared in the zero-G environment. The errors induced in the fallout limited terrestrial laboratory may then be determined for both instruments at once. Emphasis will be placed on obtaining duplicate nucleus sizes in the zero-G and 1-G environments; however, as long as accurate size distribution analyses are provided, there should be no special difficulties in interpreting the data.

With respect to special problem areas; additional work must be performed to develop adequate methods of particle generation. For the proposed tests, it will be important to use materials of only the highest purity, particularly when testing the nucleation efficiency of seeding agents such as AgI or NaCl. Additional work is also required to develop methods of generating contamination free aerosols. This work is particularly important if comparisons are to be made between theoretical predictions of particle activity and experimental observations.

#### S. 7.2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below:

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	4
Size-droplet	
Type	2
Pollutant	
Pressure	
Temperature	3
Relative humidity	5
Charge	
Rate of cooling	
Time	6
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	3
Velocity	
Liquid-water content	
Surface tension	



<u>Parameters</u>	<u>Variations</u>
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	2
Spin rate	

## S.8 PROCEDURE

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure S-1).

## S.9 REFERENCES

1. Fitzgerald, J. W., 1970: Non-Steady Supersaturations in Thermal Diffusion Chambers, J. Atmos. Sci. 27.
2. Langsdorf, A., 1936: A Continuously Sensitive Cloud Chamber, Phys. Rev. 49
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4. Squires, P., 1971: Keynote Address, the Second International Workshop on Ice and Condensation Nuclei. Dept. Atmos. Sci. Colo. State Univ. Rept.
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6. Twomey, S., 1963: Measurements of Natural Cloud Nuclei, J. Res. Atmos., 11.
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# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>Equipment set-up and checkout</li> </ul> <p>During this initial phase of the experiment, all equipment is readied for use. The SDC or CFD chambers are set up, regulators and flowmeters checked out, monodisperse aerosol generator prepared, and conditioning chamber examined for leaks or damage. All electrical service and camera equipment is examined and tested.</p>	30
<ul style="list-style-type: none"> <li>Purge SDC, conditioning chamber, all duct work, coagulation tube (if used) and electrical aerosol analyzer with dry nitrogen or filtered air.</li> </ul>	5
<ul style="list-style-type: none"> <li>Generate nuclei to be tested               <ul style="list-style-type: none"> <li>- Use will be made of a monodisperse aerosol generator or heated wire technique.</li> <li>- If NaCl nuclei are to be used, choose solution strength desired, generate droplets and allow to evaporate in conditioning chamber - the salinity of the solution and droplet size will determine the size nucleus that is produced.</li> </ul> </li> </ul>	10
<ul style="list-style-type: none"> <li>Set supersaturation (RH) - while nuclei are being generated adjust SDC to desired S%.</li> </ul>	<2
<ul style="list-style-type: none"> <li>Introduce nuclei into SDC (and EAA) and observe concentration of nuclei activated to droplet growth - Record data - Do 5 times per saturation - If sufficient nuclei remain in conditioning chamber, flush SDC, reset to new supersaturation and introduce nuclei into SDC (and EAA) and observe concentration of nuclei activated to droplet growth - Record data - Do 5 times per saturation.</li> </ul>	2-5
<p>Recycle using at least 4 different supersaturations.</p>	
<p>When complete purge SDC, conditioning chamber, all duct work, coagulation tube (if used) and electrical aerosol analyzer with dry nitrogen or filtered air.</p>	1 min to flush +5
<p>Recycle with other materials and supersaturations, at least three and as many as six standard nuclei to be used.</p>	

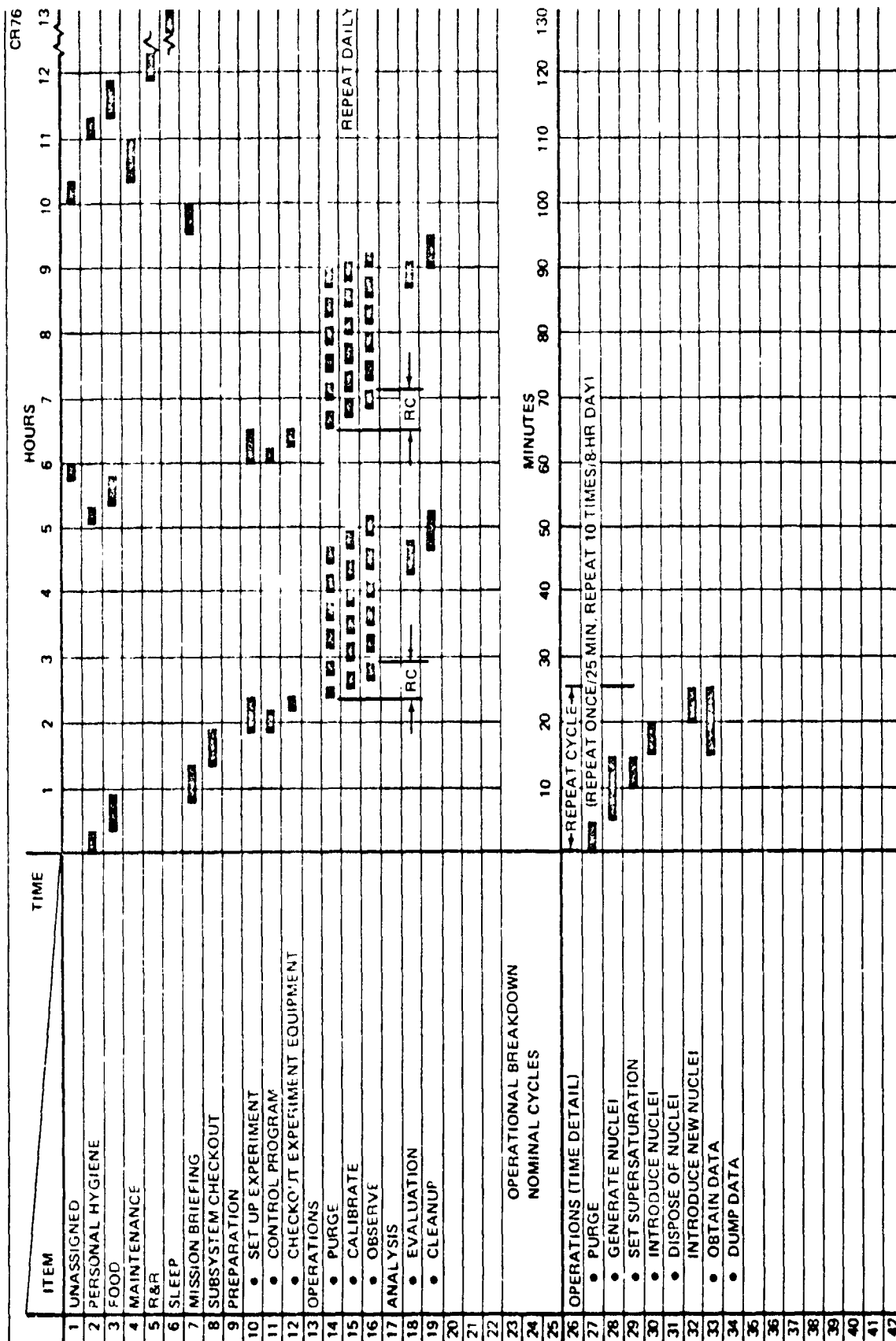


Figure S-1. Activity Timeline (One Day) Experiment Class 19

Appendix T  
CLASS 20  
UNVENTILATED DROPLET DIFFUSION COEFFICIENTS

T.1 UNVENTILATED DROPLET DIFFUSION COEFFICIENTS APPROACH 1

T.1.1 Introduction

The equation for evaporation or growth of droplets is given by Byers (1965) as:

$$r^2 = r_0^2 + (S - 1) (1 + 0.22 F R_e^{1/2}) \Delta t / (A + B) \quad (1)$$

where  $R_e$  is the Reynolds number,  $F$  is the ventilation coefficient,  $r$  is the radius of the droplet,  $S$  is the ratio of the vapor pressure to the saturation vapor pressure in the environment,  $A$  and  $B$  are functions of temperature and pressure. Their dependence on ventilation is slight, and will be neglected. For a unventilated droplet such as a droplet falling or being supported in a wind tunnel, the ventilation factor:

$$f = 1 + F 0.22 R_e^{1/2}$$

reduces to 1.

The  $f$  factor was first investigated by Frössling (1936). Kinzer and Gunn (1951) and Beard and Pruppacher (1971) have also studied evaporation of <<pure>> water droplets in the size range 140-40  $\mu$  in diameter. In the first instance the drops were in free fall at 50 percent relative humidity. The droplets were charged and sensed by induction. Beard and Pruppacher (1971) suspended drops in a wind tunnel and related the terminal velocity of the drop to the velocity of the air stream. In each case the authors were able to obtain the ventilation factor ( $F$ ). The ventilation factor has also been examined theoretically by Abraham (1968) and Woo and Hamielec (1971).

As a further check, it is desirable to ascertain the unventilated diffusive evaporation rate. This can be used as a base for the wind tunnel experiments on small droplets. At the present time, no unventilated evaporation rate data are available except for droplets supported on spider webs, etc.

These data will fill a gap in our knowledge concerning the evaporative behavior of small droplets (the size of fog drops) and those droplets which are smaller than ones which may be readily observed, unsupported in a terrestrial laboratory.

This class of experiments on pure water is scientifically interesting but not of nearly as great an importance to man as experimentation with drops coated with mono-layers or organic long chain molecules. Fuchs (1964) suggested that the persistence of fog near urban areas was in some way related to pollution. Hoffer and Mallen (1970) have shown that drops coated with automobile exhaust residue, plasticizers and the long chain alcohols (cetyl alcohol) do inhibit evaporation. Garrett (1973) has summarized the results from several materials, principally long chain molecules. The time dependence of the coating, the length of time the coating or mono-layer should exist prior to sublimating is a parameter in great doubt at the present time. Warner (1970) has calculated the equilibrium for cetyl alcohol and finds the time to be not excessive. However, the time is in excess of that period droplets can be supported in a terrestrial wind tunnel.

Recently, Fitzgerald (1974) has computed that there is a sufficiency of organic material to retard evaporation over the North Sea. However, the organic compounds are not identified. The correctness of this approach towards stabilizing unsaturated fog is still questionable since the organics have not been tried for their effectiveness.

The residual drop size after a fog evaporates is of importance for several reasons: (a) larger particles will constitute a greater obstruction to visibility; (b) larger particles will interact with the radiative transfer processes differently; and (c) the collection and retention processes of plants and animals will be effected.

The question of the surface area coverage provided by the mono-layer is another prime factor. How much area must be covered may be a function of the Reynolds number which governs the flow around the drop. These interactions need to be studied as they reflect the total amount of organic material necessary to retard evaporation in a fog.

#### T.1.2 Objectives

The experimental objectives are as follows:

- a) Delineate a standard evaporation rate for small drop experiments.
- b) Provide data on the effectiveness of various naturally occurring and artificial materials in inhibiting evaporation.
- c) Provide information on the time history of an evaporating drop to determine how long the pollution mono-layer is effective.
- d) Provide data on the percent surface area coverage versus evaporation rate to determine the amount of material and its distribution on the drop needed to inhibit evaporation.

#### T.1.3 Scientific Justification

Currently our total knowledge of evaporative behavior in fog and cloud is small. Thus the data can lead to a better understanding of the fog and cloud phenomena which undoubtedly will become a more important part of the terrestrial research program.

The above objectives also bear on the stability of a "polluted" fog. The evaporation time of the droplets will radically change when the air becomes subsaturated if a mono-layer is present. This affects the lifetime of the fog. This in turn affects attempts at fog modification and forecasting the time of fog burn off.

Fog is a complex problem and costs many man hours/year. The knowledge of the droplet lifetime in a polluted environment (subsaturations and time required prior to evaporation) will aid in understanding the dissipation of fog. The evaporative behavior is important in urban areas where the ingestion of harmful pollutants may become important.

The growth of droplets can be investigated in future experiments to be absolutely certain that mono-layers do not inhibit growth.

#### T.1.4 Applications

The information can be directly applied towards:

- a) dissipation of fog,
- b) stabilization of fog and clouds, and
- c) the application of organics to water and droplets to inhibit evaporation.

#### T.1.5 Terrestrial Laboratory Limitations

The limitations in a terrestrial laboratory are:

- a) Droplets must be supported on a thin strand of spider web, plastic or other material which does not conduct heat. The drop is still partially distorted and the effect of this distortion must be considered. It is difficult to obtain the mass of such drops if the size is small without, in general, radically altering the experimental field.
- b) Ventilating small supported droplets causes oscillation and other viewing problems.
- c) Observations in wind tunnels of coated or uncoated droplets permit viewing the droplet only at the Reynolds number ( $R_e = \frac{2rv}{\eta}$ ) at which the droplet can be supported. It is impossible to obtain the unventilated case in this type of experiment.
- d) The observational difficulties are extreme with small drops, those of cloud and fog droplet size.
- e) The mono-layer on the droplet is almost impossible to observe in the wind tunnel system due to the restrictions on disturbing the flow around the droplet and the fact that the droplet is fluctuating rapidly.
- f) With the exception of the relatively unrealistic strand supported droplet, it is not possible to model the Reynolds number, i. e., use a larger drop with a smaller velocity.

#### **T.1.6 Zero-Gravity Opportunities**

Experimentation under zero g permits:

- a) Unventilated evaporation determination in a realistic environment.
- b) Observation of the surface area of the drops which is covered when evaporation is inhibited. This can be accomplished as a function of Reynolds number. A large enough drop (200  $\mu\text{m}$  diameter) can be used and the Reynolds number modelled since the flow around the drop is of vital importance.
- c) The experiments on mono-layers can be conducted for sufficient lengths of time to permit an approach to final equilibrium.

Data on the stabilization of droplets can be obtained under zero g conditions that would take a much longer time to obtain under terrestrial conditions. A reasonable estimate might be a period of ten to fifteen years.

#### **T.1.7 Approach**

##### **T.1.7.1 General**

The approach to the droplet evaporation problem has been, in the first investigations, to support the droplets on strands. These were not highly successful.

Kinzer and Gunn (1951) used various techniques but primarily freely falling drops. Beard and Pruppacher (1970) and Hoffer and Bowen (1973) used wind tunnels. The evaporative behavior of droplets with some inhibitors has been studied by Garrett (1973) and Hoffer and Mallen (1970). Garrett used drops supported on a fiber while Hoffer and Mallen (1970) used a wind tunnel. Hoffer and Bowen (1973) also studied the effects of various vapors on evaporation in a wind tunnel.

The great advance that zero g makes is that the drop can be stopped and examined for the extent or absence of a mono-layer prior to evaporation. The drop may then evaporate either in motion or stationary. If the droplet is in motion, the Reynolds number can be varied by changing the drop



velocity. The drop could be put into a circular path inside the chamber. Since there will be drag on the drop, a means of providing a force to keep the drop in motion will be necessary.

The procedure for the series of experiments on stationary droplets would be:

- a) Humidify the general chamber to the lowest dew point temperature desired by evaporating water into the chamber using a syringe with a heated needle. The dew point would be measured intermittently with a ruggedized Cambridge Dew Point Hygrometer.
- b) Generate a droplet and position it so that it can be observed with the microscope and digital TV camera (simultaneously mirrored with a beam splitter) so that observation may take place with data acquisition. The surface temperature of the drop should be observed since evaporating drops approach the wet bulb temperature as an equilibrium.
- c) Let the droplet evaporate for a period of time from 200 sec. to 2,000 sec. and observe the  $T$ ,  $T_{\text{drop}}$ ,  $T_{\text{dew point}}$  pressure, and drop size.
- d) Repeat the procedure for pure water and drops that have a partial mono-layer at the outset. The areal extent of the mono-layer should be determined immediately after drop generation. The inhibiting agents (such as cetyl alcohol) might be applied to the droplet by coating the meniscus with a solution. Subsequent penetration of the meniscus by the drop generating rod would carry some agent onto the surface. Evaporation inhibiting agents, both natural and artificial, will be used.

The procedure in the case of the ventilated droplets would be the same for steps a, b, and d. In step c, the drop would evaporate while being ventilated. The lower Reynolds number regime from 0.5 to 20 would be modelled by starting with a 200  $\mu\text{m}$  radius drop, whose velocity at 20°C would be given by

$$v = \frac{\eta R_e}{2r} \quad (2)$$

A variation from the wet bulb towards warmer temperature when the drop is evaporating would indicate the effect of the mono-layer.

It is obvious that the droplet can be put into a circular orbit with, say, a radius of .02 meters. A constant acoustical force could be applied to keep the angular velocity constant. This would mean a slightly changing Reynolds number.

The pure water droplets and the droplets with a measured amount of surface agent would evaporate under the controlled conditions for times between 200 and 6,000 seconds dependent upon the final radius of the droplet.

The Reticon would be used to measure the droplet size as well as the droplet velocity (the droplet velocity obtained by strobing). A variable magnification would be provided by the optics in the stereo microscope. A beam splitter would deflect the image into the Reticon camera. It is anticipated that some advance in the state of the art would occur but even if it does not a magnification of ten will be adequate for a resolution of 10  $\mu$ m in diameter.

This experimental procedure will delineate the evaporation of drops for pure water (to serve as a reference) and for drops which are surface contaminated. The answers are of fundamental importance to fog and smog stabilization and thereby to the health of mankind.

The velocity of the 200  $\mu$ m drop to model various Reynolds numbers is given in Table T-1.

Table T-1  
REYNOLDS NUMBER VERSUS VELOCITY FOR A 200  $\mu$ m  
DIAMETER DROPLET

Re	0.5	1	2	4	8	16	
v	.153	.075	.038	.019	.0095	.0048	meters/sec.
r	200 $\mu$ m	200 $\mu$ m	200 $\mu$ m	200 $\mu$ m	200 $\mu$ m	200 $\mu$ m	

#### **T. 1. 7. 2 Experiment Parameters**

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	1
Type	2
Pollutant	4
Pressure	1
Temperature	3
Relative humidity	3
Charge	
Rate of cooling	
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	5
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### T. 1.8 Procedure

General activity details are given below in a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figure T-1).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
<ul style="list-style-type: none"> <li>• Purge chamber</li> </ul>	2
<ul style="list-style-type: none"> <li>• Reduce pressure to 500 mb. Stabilize temp. at 20°C</li> </ul>	1/2
<ul style="list-style-type: none"> <li>• Humidify (50 percent) (85 percent) (90 percent)</li> </ul>	5
<ul style="list-style-type: none"> <li>• Generate and position drop</li> </ul>	1/6
<ul style="list-style-type: none"> <li>• Do: A. Coat with monolayer or B. Leave alone.</li> </ul>	
<ul style="list-style-type: none"> <li>• Drop evaporates to 30µm or for 30 minutes.</li> </ul>	5 (Aver)
<ul style="list-style-type: none"> <li>• If <math>\Delta m / \Delta t</math> changes. Note extent of monolayer (microscope) at period end. Push drop aside.</li> </ul>	
<ul style="list-style-type: none"> <li>• Repeat pure water and monolayers (ventilated and unventilated) for 10 drops each condition. Then return to initial.</li> </ul>	

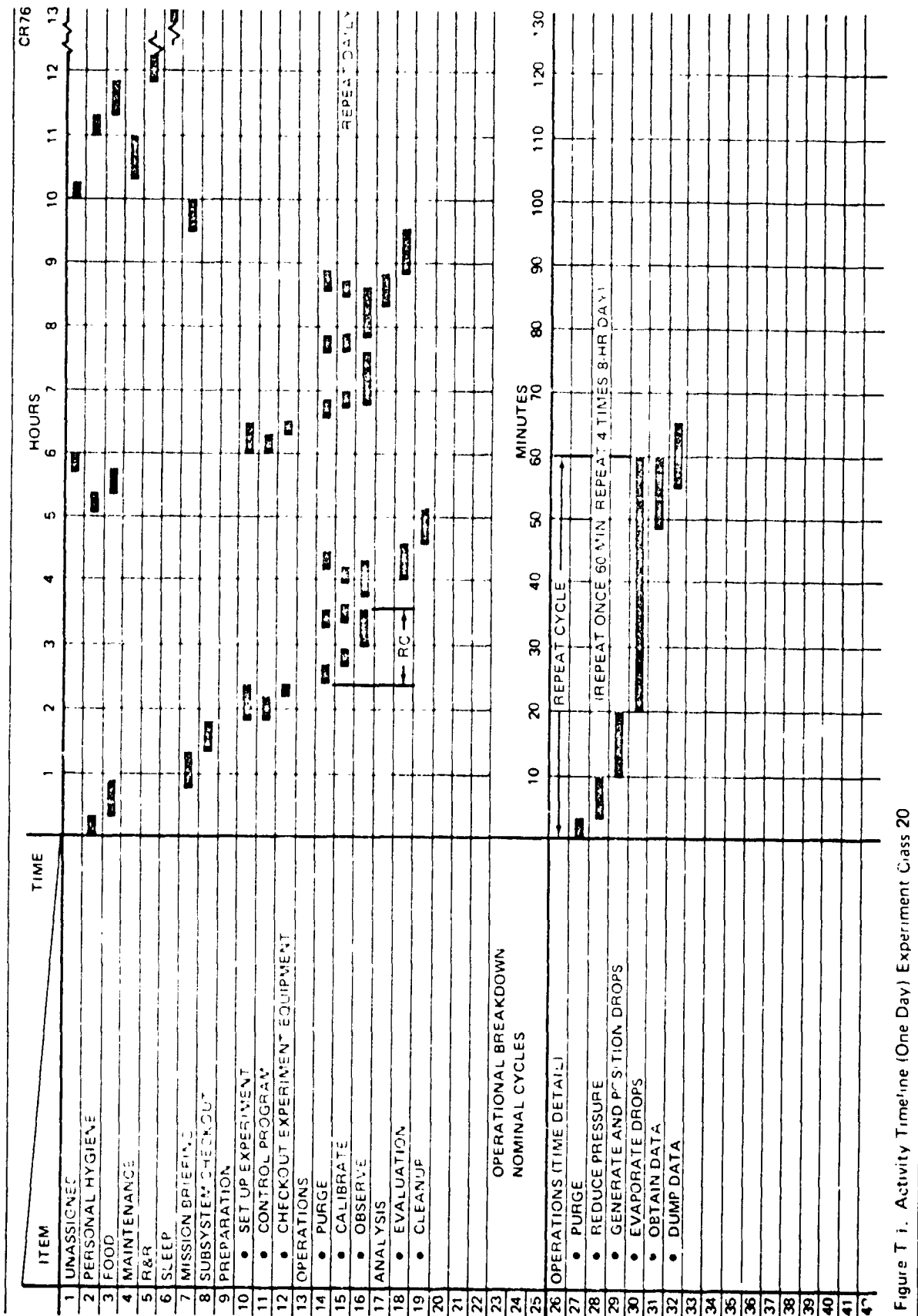


Figure T i. Activity Timeline (One Day) Experiment Class 20

#### T.1.9 Conclusions

The experiment is relatively simple requiring uncomplicated procedures and controls. It is of great value to the understanding of the surface aerosol and its effect on plants and animals. The results will also aid our understanding of the problems of fog dispersal. In addition, the modelling of the Reynolds number permits observation of the amount of surface agent needed to inhibit evaporation. The zero gravity concept is essential to the conduct of the latter part of the experiment.

The total time required is seventy five hours which should be available within the experimental period of one week.

#### T.1.10 References

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## 5.2 UNVENTILATED DROPLET DIFFUSION COEFFICIENTS APPROACH 2

### T.2.1 Introduction

The diffusive growth of cloud droplets is of general meteorological importance. Droplet growth theory exists in a wide variety of theoretical treatments. It might be inferred from the scores of applications of the theory that it is thought to be in pretty good shape, as long as growth is not controlled to any great extent by either surface or ventilation effects. The diffusive growth theory tends not to predict the observed drop spectra (Fitzgerald, 1970, offers a possible exception) and is blamed on factors other than the possible inadequacy of the theory itself. For instance, the theory leaves out non-uniform gas effects and treats the diffusion processes as if they were taking place in a solid that is immune to thermal expansion. Quite aside from factors such as surface kinetics and ventilation, it would be reassuring to witness a convincing experimental test of the usual theory (Mason, 1971 see page 122) which postulates mass and heat transport controlled completely by molecular diffusion and conduction under conditions where gravity induced effects such as ventilation are absent. The one might zero in on any real theoretical deficiencies.

The zero-g environment lends itself to such an experimental study of drop growth, particularly when surface effects exert a controlling influence. Rooth (1957) identified the role of surface kinetics on vapor condensation and parameterized the effect by means of a sticking coefficient (hereafter called  $\beta$ ). If  $\beta$  is sufficiently small, it can cause meteorologically significant changes in drop growth rates. A similar coefficient is associated with heat transfer, namely the thermal accommodation coefficient,  $\alpha$ . Carstens et al (1974) have discussed the possible role of both  $\alpha$  and  $\beta$ . While Alty and Mackay's (1935) values of  $\alpha$  and  $\beta$  (1 and 0.036) are often used in meteorological applications, the general literature includes values of  $\beta$  ranging from 0.01 to 1.0 (Mills and Seban, 1967) which places the subject in utter confusion. (Values of  $\alpha$  are frequently assumed to be unity for lack of better insight into the matter, if  $\alpha$  is included at all in the analysis.) Since the spread in  $\beta$  includes values of  $\beta$  sufficiently small to be important in atmospheric analysis, one must regard its measurement as important. It should finally be mentioned that the recent measurements of Vietti and

Schuster (1973), made at much greater supersaturations than occur in the atmosphere, imply low values of  $\alpha$  and  $\beta$  (0.1 and 0.035 respectively) using the "conventional" theory (Fukuta and Walter, 1970; Carstens and Kassner, 1968). Problems in these measurements exist, however, that could be alleviated in a zero-g environment. It would be nice to be able to positively separate the ventilation factor from the problem of determining appropriate values for  $\alpha$  and  $\beta$ ; zero-g offers this opportunity. Moreover, it is difficult to study the diffusional growth of larger droplets in the terrestrial laboratory because of their high terminal velocities.

#### T. 2. 2 Objective

The objective of this experiment is to determine the growth rate of droplets from about  $1\mu$  to several tens of microns. Identify the effective (see Approach) diffusion coefficient and to measure the composite effects of sticking coefficient and thermal accommodation coefficient (hereinafter called "surface effects"). Attempt to distinguish the separate contribution of the surface effects and try to evaluate the role of various contaminants, e. g., surfactants, in the overall growth process.

#### T. 2. 3 Scientific Justification

Several aspects of drop growth enter into problems dealing with the modeling of atmospheric processes where a phase change from vapor or liquid or liquid to solid (in cases where a spherical particle can be assumed) is involved. In the development of clouds and fogs, droplets grow to various sizes via a growth law that incorporates various physical and chemical mechanisms. The growth law dictates growth rates, final sizes, and size distribution. Generally the droplets are assumed to be noninteracting, except through the use of a cell model which takes into account the decay of supersaturation some distance from the droplets. In the atmosphere, two droplets would not remain in close proximity for very long periods of time because slight differences in their size results in differences in their fall velocity which would separate the pair rather promptly compared to the speed of growth. In the absence of gravity, the statistical distribution of closely spaced pairs on the final drop size distribution would be likely to cause much larger effects and would need to be properly taken into account in a zero-gravity experiment. Such effects can be estimated if the total drop population is known (Williams and Carsens, 1971; Carstens, Williams, and Zung, 1970). On the other hand, ventilation

effects would be notably absent so that the assumption of spherically-symmetric diffusion fields would be expected to be an accurate representation.

In non-freezing precipitation situations, drop growth by direct condensation sets the stage, so to speak, for the collision-coalescence process.

Condensational growth plays a role in the size distribution of fogs, which affects visibility. In cloud or fog modification, deliberate or inadvertent, the way condensational growth "sets the stage", i. e., determines the initial size distribution, can depend on factors, such as surfactant and/or soluble contaminants, which enter into the growth law. Present droplet growth theory, is sufficiently well parameterized to allow adequately for the incorporation of these effects; however, we have not nearly settled what values these parameters should have for pure drops. The situation for high and low droplet growth rates is quite likely to be different. Water is a very active surface and becomes contaminated quite rapidly. Slow growth rates might enhance these effects. It is difficult to see how one can expect to predict through numerical modeling the effectiveness of various modification processes with the present gap in knowledge.

Detailed Theoretical Reasoning: The drop size spectrum, itself an important microphysical attribute of a cloud, is intimately related to cloud microphysical phenomena as well as cloud dynamics. The zero-g program applied to the study of microphysical phenomena, is appropriate to describe those specific processes which contribute to the spectral size distribution, and which are amenable to study in the zero-g environment. Such questions are of course tied up with the way droplets grow and/or evaporate.

The equations describing growth (or evaporation) can be written down in a form familiar to atmospheric scientists (Carstens et al., 1974).

$$(a + l) \frac{da}{dt} = \frac{[S - S_{sat}(a)] \rho_{eq}(\infty)}{\frac{1}{D} + \frac{bL}{K}} \quad (1)$$

where

$$I = \frac{(\lambda_v/\beta) \frac{1}{D} + (\lambda_g/a) \frac{bL}{K}}{\frac{1}{D} + \frac{bL}{K}}$$

Here  $D$  is the diffusion coefficient of water vapor in air,  $K$  the thermal conductivity of the air-vapor mixture,  $L$  the latent heat of condensation,  $S$  the applied supersaturation ratio,  $\rho_{eq}(\infty)$  the equilibrium vapor density far from the drop,  $b$  the slope  $(\Delta p/\Delta T)$  of the equilibrium vapor density versus temperature curve,  $\lambda_v$  and  $\lambda_g$  approximate mean free paths referring to the vapor and air,  $a$  is the thermal accommodation coefficient,  $\beta$  the condensation coefficient,  $a$  the drop radius,  $t$  the time, and  $S_{sat}(a)$  the equilibrium saturation which we will take to be the usual Köhler result,

$$S_{sat}(a) = 1 + \frac{r^*}{a} - \frac{A}{a^3} \quad (2)$$

with  $r^*$  denoting the effects of drop curvature, and  $A$  solubility of salt.

It is well known that this growth law implies two (purely microphysical) mechanisms which can lead to a broadening of the radial size distribution. This implication can be clarified in the following argument. Consider two isolated drops of radius  $a_1$  and  $a_2$ , where  $a_1 > a_2$ , which are growing under identical ambient conditions. Equation (1) applied to both drops, and the two equations when divided yield:

$$\frac{da_1/dt}{da_2/dt} = \left( \frac{a_2 + I}{a_1 + I} \right) \left( \frac{1 - \frac{r^*}{s-1/a_1}}{1 - \frac{r^*}{s-1/a_2}} \right) \quad (3)$$

where we are considering drops which are well activated, and have therefore dropped the solubility term. We can rewrite Eq. (3) so as to reveal three separate tendencies:

$$\frac{da_1/dt}{da_2/dt} = \left( \frac{a_2}{a_1} \right) \left( \frac{1 + I/a_2}{1 + I/a_1} \right) \left( \frac{1 - \frac{r^*}{s-1/a_1}}{1 - \frac{r^*}{s-1/a_2}} \right) \quad (4)$$

The first factor implies a narrowing of the radial size distribution. This term dominates at large radii ( $S > 1$ ) and is responsible for the narrowing process commonly ascribed to diffusion. The second term, which through  $l$  is associated solely with surface kinetics, implies a broadening. Regardless of the magnitude of  $l$ , it is clear (from Eq. (3)) that this broadening can never "overtake" the narrowing tendency of the first term, so the surface kinetic effects tend only to retard the narrowing tendency of diffusive growth, they cannot reverse it. The broadening effect implied by the third factor is due to drop curvature, and clearly it can be magnified at low supersaturation ratios. This effect is well known (Maser, 1972) and is usually thought to be too sluggish a process (Elton et al, 1958) to significantly broaden the spectrum.

Of the two processes identified above surface kinetics represents certainly the most difficult to elucidate, since surface effects generally involve complex and poorly understood processes. On the other hand since the parameterization of this process is so simple (changing  $a$  to  $a + l$ ) it is a comparatively straightforward matter to establish, or at least estimate, values of  $l$  which could lead to significant cloud physical effects. Rooth (1957), Warner (1969), for example, have indicated values for  $l$ , or rather  $\beta$ , that would be significant. Because of the complexities involved it is worthwhile to augment the discussion above and extend (and correct somewhat) the arguments of Rooth.

Surface Kinetic Effects: It has already been pointed out that surface kinetic effects retard the narrowing tendency of purely diffusive growth, and thus generally deform the drop size spectrum in a direction that has the possibility at least of leading to agreement with observation. It is clear that the greater the surface control of the transport process, the more pronounced is this retardation. Likewise the lower  $\alpha$  and/or  $\beta$ , the more important it is to know their magnitudes accurately. It may be said (roughly) that if  $l$  is less than about 3 microns, its effect is not sufficiently significant to justify whatever effort is required to determine  $\alpha$  and  $\beta$ .

The most obvious questions is what are the values of  $\alpha$  and  $\beta$  for pure water? The answer (evidently) is that values of  $\beta$ , the condensation coefficient, have been measured which run from about 0.01 to unity;  $\alpha$  is usually taken at, or

near, unity. Rohsenow and Sukhatme (1964) suggests that  $\beta$  is probably equal or very close to unity, but that with traces of impurities  $\beta$  can take on values much less than unity. Mills and Seban (1967), who subscribe to this view, have listed an array of values measured by various investigators. They divide this array into two groups: (1) those who measure low values ( $\beta < 0.4$ ), and (2) those who measure high values ( $\beta > 0.4$ ). Usually, if the coefficients are used at all by cloud physicists, they use Alty and Mackay's (1938) values of  $\alpha = 1$ ,  $\beta = 0.036$ . Warner (1969) and Fitzgerald (1970) are notable exceptions in that they contemplate other values in order to secure agreement with their observations.

Three very recent works (Sinnarwalla and Alofs, 1974; Chodes et al, 1974; Carstens and Carter, 1974) have suggested that low values of  $\alpha$  and  $\beta$  are appropriate for dropwise condensation in air. In these works, the drop growth process was "fit" by choosing  $l$  in the theory described by equation (1). The results are expressed by assuming  $\alpha = 1$  and allowing  $\beta$  to take on a value which will give the desired  $l$ . Chodes et al arrive at a value of  $\beta = 0.033 \pm 0.005$ . While this value is low enough to be non-negligible in drops size calculations, Chodes et al conclude that it is not sufficiently small to fully explain the cloud droplet size distribution of Warner (1969). Sinnarwalla and Alofs however find an average value of  $\beta = 0.024$  which is in excellent agreement with the value ( $\beta = 0.023$ ) determined by Carter and Carstens (1974) at much higher supersaturations for drop growth in "pure (nuclei free) air. Sinnarwalla and Alof's observation indicates that "there is more than one value of  $\beta$  in the atmosphere." Their data implies a range of  $0.01 < \beta < 0.1$ .

In view of the data to date, it would appear, for dropwise condensation in the atmosphere, that either  $\alpha$  or  $\beta$ —presumably  $\beta$ —is low enough to be important in cloud physics. Granting this, several problems need to be resolved. The first problem, which has already been outlined for the zero-g experiments, consists in the determination of  $\alpha$  and  $\beta$  separately. This is important because the single parameter  $l$  represents a weighted average of  $\alpha$  and  $\beta$ , as indicated by its definition below Eq (1). The weighting of  $\alpha$  is strongly temperature dependent so that while the effects of  $\alpha$  and  $\beta$  on  $l$  are comparable

at 0°C, at 25°C the increased thermal weighing causes  $\alpha$  to be the dominant factor. Thus  $l$  has a different temperature dependence, depending on whether it is  $\alpha$  or  $\beta$  that is small. If  $\beta$  is small, as generally assumed on the basis of Alty and Mackay's measurements,  $l$  will have more effect at lower temperatures than at high.

Aside from the way  $\beta$  is "weighted into" the growth equation, there is the question of variable  $\beta$ . If  $\beta$  (or  $\alpha$ ) is affected by contaminants it is naive to suppose that it can be properly characterized by a single value. An example is afforded by organic surfactants. Hughes and Stampfer (1970) have shown drops partially covered with surfactants exhibit enhanced evaporation rates. Since the atmosphere contains surfactant material, it is reasonable to suppose it to be distributed among cloud droplets, leading to a distribution of sticking coefficients. In addition,  $\alpha$  will be radially dependent since whatever surfactants are on the drop will tend to be diluted as it grows. Finally, the condensation coefficient may also change due to the "aging" of the droplet. This could be brought about by the continuous scavenging of various contaminants as well as the rate at which they become distributed and oriented throughout the volume and surface of the droplet.

Finally it should be mentioned that surface kinetics affect drop temperature. Since  $\alpha$  and  $\beta$  (thermal accommodation and sticking coefficients) control the transport process to an extent dictated both by the radius of the drop and the strength of the inequalities  $\alpha \ll 1$  and  $\beta \ll 1$ , they would also be expected to play a role in the determination of drop temperature. The temperature field characterizing a droplet and its surroundings is known to be discontinuous at the drop surface (Carstens and Kassner, 1968). There is thus a temperature jump at the surface:

$$T_J = T_D - T_a$$

where  $T_J$  is the jump,  $T_D$  the drop temperature, and  $T_a$  the gas temperature just outside the drop. It is easy to show that

$$T_J = \frac{a}{l_a} \Delta T_a$$

where

$$\Delta T_a = T_a - T_\infty = \frac{L}{K} \frac{\rho_{eq}(\infty) (S - S_{sat})}{1 + \frac{l}{a}}$$

Clearly  $T_J$  goes to zero as the radius becomes large;  $T_J$  is evidently a manifestation solely of thermal controlled surface transport.  $\Delta T_a$ , on the other hand, is clearly a product of a purely diffusive term and a surface term. The drop temperature itself is

$$T_D = T_\infty + \Delta T_a \left(1 + \frac{l}{a}\right)$$

it is clear from the above analysis that for sufficiently small droplets, the drop temperature depends upon the thermal accommodation coefficient.

The Curvature Effect: "Ripening". The broadening effect due to curvature is as a phenomenon distinct from surface broadening, but not entirely independent of it. The broader the spectrum, the more likely it is that ripening can take place; a broad spectrum ought to enhance the ripening rate somewhat as well.

The nature of the ripening process can be illustrated very simply by using the Köhler curve and considering an artificially simple distribution consisting of two families of drop  $a_1$  and  $a_2$ ,  $a_1 > a_2$ . Suppose further that the initial radii of these two drops are the equilibrium radii at 100% relative humidity, and that an initial, undriven supersaturation exists which is sufficient to activate each drop (see Figure T-2).

The final equilibrium configuration ought to be  $a_2$  at some stable equilibrium and  $a_1$  at an unstable equilibrium, as shown by  $a_{1F}$  and  $a_{2F}$ . Such a configuration will be established only after a long period of time. There should be an intermediate "stagnation" period where the droplets radii are comparable; separation will occur when the supersaturation cuts between (see Figure T-3) the droplets. The evolution of this process is displayed in the following



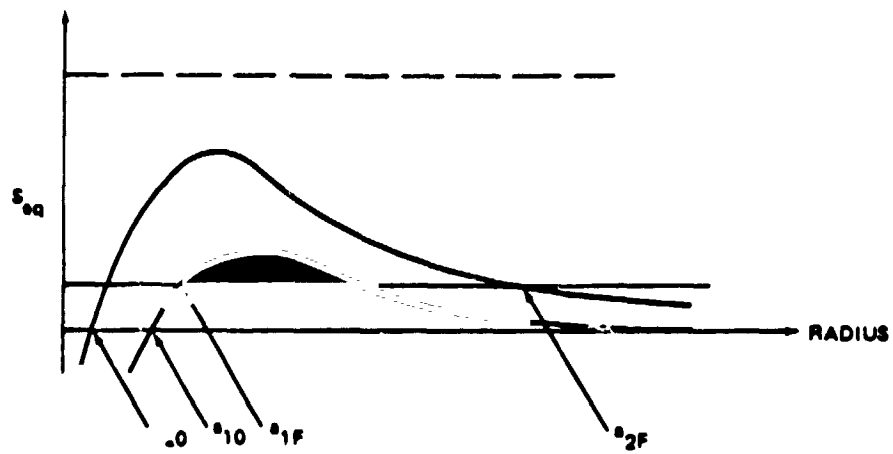


Figure T-2. Kohler Curve

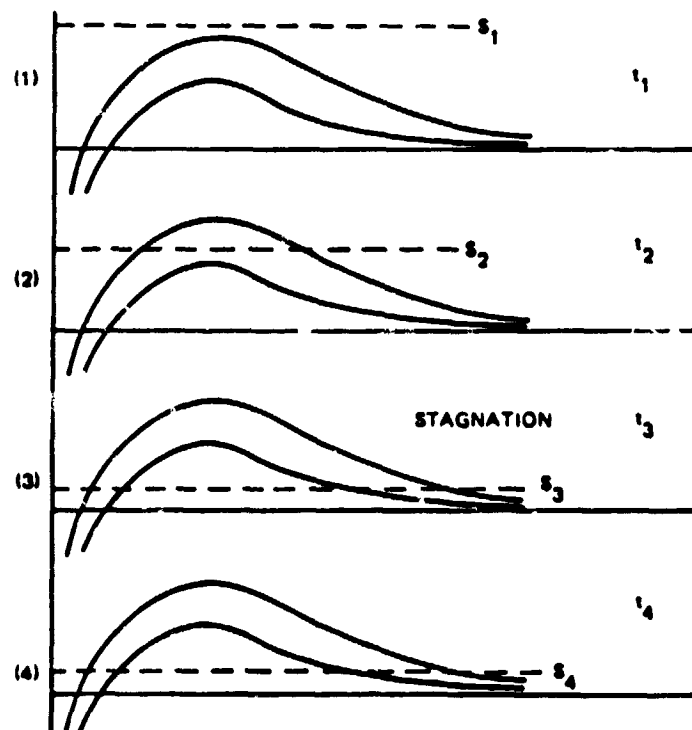


Figure T-3. The Curvature Effect Evolution

4 figures for various times,  $t_1$ ,  $t_2$ ,  $t_3$  etc. ( $t = 0$  shown above). The supersaturation retreats as a result of droplet growth as  $t$  increases from  $t_1$  to  $t_4$ .

The central question regarding the ripening process is how long does the stagnation period last. Mason (1973), following Elton et al (1958), suggests that this time is too long to be significant, at least for growth. Preliminary computer calculations, however, suggest that evaporation to equilibrium (inactivated) radii may take place in reasonable times. This of course represents a broadening of the spectrum.

In not driving the supersaturation we are in a sense leaving out dynamic effects; here is a case where certain downward fluctuations in supersaturations, brought about presumably by certain parcel dynamics, could enhance the ripening process. Zero-g would provide enough stagnation time to observe the ripening process clearly.

#### T.2.4 Applications

There are a wide range of applications of diffusional droplet growth theory. Droplet growth is an important consideration in the operation of steam turbines in power plants where it is essential that droplets do not grow to sufficient size that they cause erosion of the turbine blades. Droplet growth on surfaces determines the efficiency of operation of fractional distillation systems such as those which process petroleum products. The desalination of salt water by distillation also relies on the same fundamental principles being studied here.

Because this is a cloud physics program, the most conspicuous applications occur in connection with various aspects of atmospheric science. In cloud formation, the initial drop size distribution is determined primarily by the interplay between the diffusional droplet growth mechanism and the spectrum of activities associated with the cloud condensation nuclei. What happens during the condensation stage of cloud development sets the stage for later events such as turbulent mixing and collision coalescence. Precisely what happens and when, is determined by just how large droplets can grow in the time allotted by the diffusion mechanism. Thus, a knowledge of the factors

which affect the observed rate of growth of droplets and the development of an analytical formulation, which accurately represents the growth rate of cloud droplets over a broad size range, a broad range of rates, a wide range of temperatures and taking into account the effect of different types of impurities - both ionic solutes and surface contaminants, are high on the priority list of things which need to be firmly understood in order to meet the more exacting needs of cloud modelers. Such an analytical formulation would allow the assessment of various cloud and fog modification schemes.

#### T. 2. 5 Terrestrial Laboratory Limitations

The general limitations of terrestrial experiments may be attributed directly to (1) drop fall, and (2) free convective currents. These have the major consequence that a limited observation time is imposed on the experiments due to droplets falling out of the region of observation. Moreover, the droplets are disturbed by extraneous convective currents arising from temperature differences in the chamber and the accompanying differences in density. When such limitations are removed by the introduction of a support fiber or platform, the growth of the droplet is interfered with through contamination from the support, thermal conduction through the support and surface diffusion of molecules over the surface of the support.

The way in which gravity limits experimental investigation in two commonly employed experimental set-ups are indicated. When drop fall under gravity is used to measure growth or evaporation rates (Duguid and Stampfer, 1971), a limitation was imposed in that the residence time of individual droplets was such that only one datum could be extracted from each drop. Duguid and Stampfer attributed their scatter in data to the fact that they had to examine a number of individual drops and obtain a statistical average. The different histories of these drops, formed on different nuclei, they felt, caused the scatter in their data. Clearly it would be helpful in these experiments to subject the same drop, or set of drops, to successive and/or prolonged measurements of evaporation and growth. Using fall rates in the measurement would obviously not apply in the zero-g situation but Mie scattering or laser scattering could still be used with excellent resolution capability as demonstrated by Vietti and Schuster (1973).

Terrestrial laser scattering has been used by Vietti and Schuster (1973) to measure growth rates at high supersaturations. A longer growth time would allow more careful identification of the initial Mie peak which is essential in order to establish the size of the drop initially. Longer observation times, especially in the expansion chamber under zero gravity, would facilitate this identification by permitting sufficient time for the drops to evaporate to a radius less than that corresponding to the first peak where repeated growth and evaporation is observed. "Missing" the first peak, or several peaks, can lead to errors in the radius that will lead to unrealistic determinations of sticking and thermal accommodation coefficients.

#### T. 2. 6 Zero-Gravity Opportunities

The removal of gravitation circumvents two basic limitations of the terrestrial laboratory facility: (1) It completely eliminates drop fall, thus providing all the advantages of a supporting platform, with none of the disadvantages such platforms introduce; (2) Free convective currents are eliminated even when slight thermal inhomogeneities exist. Both of these factors allow longer observation times which are essential to doing experiments utilizing the low supersaturations found in the real atmosphere in expansion chambers.

In addition to providing circumstances under which drop growth rates can be pinned down, measurements of large drop growth rates (at zero-g), compared with terrestrial measurements, can lead to a quantitative estimate of the ventilation factor. Since zero-g offers the possibility of a leisurely measurement at any size (using laser scattering), internally consistent runs may be utilized. That is, the parameters that affect growth at only small sizes may be eliminated by measurements carried to sufficiently large sizes (see Approach).

#### T. 2. 7 Quantification

Experiments on the growth and evaporation of single droplets and clouds of droplets is extremely important to the unraveling of several important questions in cloud physics. Experiments utilizing various means of supporting individual droplets have suffered from various problems which raise

serious questions about the validity of the results obtained. The outlook for overcoming the objections and problems associated with these support techniques is not an optimistic one. The experience of people working with poly-water is a clear cut example of the extreme difficulties involved in controlling contamination arising from surface film transport mechanisms when one is dealing with small quantities of the parent material such as is the case in most droplet growth and evaporation experiments of interest to cloud physics. The zero gravity laboratory capability eliminates the principle problems arising from droplet support techniques and will advance progress in this area dramatically.

Earth bound laboratory observations on clouds are severely limited by the observation times which can be usefully obtained before the cloud falls to the floor of the cloud chamber under gravity. The experiment time available depends on the height of the chamber and the size of the droplets to be observed. Droplets in excess of  $25\ \mu$  radius fall so rapidly that the growth and evaporation of such droplets cannot be adequately studied without resorting to some droplet support technique. Categories of experiments dealing with larger droplets such as abound in maritime clouds can probably only be adequately carried out under zero gravity conditions. Moreover, stationary droplet systems offer great observational advantages in both the detailed information that can be obtained from a given experiment and the capability of repeating growth and evaporation cycles successively on the same droplet system. The effect of gradual contamination from the gaseous environment can be effectively observed in zero-g whereas in earth bound experiments using fiber supports, the source of the contamination will always be in doubt since surface physics has not yet advanced to the point where such factors can be realistically dealt with. The zero-g laboratory should boost the ability of the cloud physics community to get a real handle on the host of surface physics problems which continue to plague a real comprehension of both cloud physics problems and a group of air pollution problems which involve the effects of both soluble contaminants and surface active contaminants on the evaporation and condensation stability of liquid droplets suspended in the atmosphere.

The general lack of definitive quantitative data from laboratory experiments is reflected in the slow growth of analytical theoretical descriptions of the phenomena which are adequate to describe the processes. Aerosol mechanics (which includes much of cloud physics) suffers generally from a real lack of effective tools which can be brought to bear on the problems of crucial interest. The advent of zero-g brings a long needed new approach to developing experiments which have the possibility of providing the kind of detailed data on a single system of droplets. To date, most of the knotty problems have been massaged with the currently available technology over and over again with only minor gains being made. Zero-g offers an opportunity to solve these problems.

#### T. 2. 8 Approach

##### T. 2. 8. 1 General

Water drops can be grown and/or evaporated in the cooled wall expansion chamber. A wide range of temperatures and super- (or under) saturations are available and should be used. Laser scattering can be used to follow growth, particularly in the early stages where surface effects can be important.

A typical run will "start" a cloud of drops at arbitrary size and evaporate them to a size sufficiently small that the Mie peaks disappear. After a time lapse, a re-expansion will cause the cloud to grow monodispersely, and the growth measured by Mie scattering. It will be necessary to identify the first Mie peak in order to properly measure the growth rate through the region where surface effects are important. As the drop size increases, surface effects will "wash out", that is they will no longer affect growth rates, and subsequent growth ought to proceed at a constant areal rate (assuming fixed supersaturation). Thus the early part of the run should show surface effects, while the later part of the run will permit a measurement of the effective diffusion coefficient (which can of course be compared with presently calculated values, since the physical coefficients involved have been measured). A subsequent re-evaporation, in addition to routinely checking for reversibility, can serve again to identify the initial Mie peak.

The effective diffusion coefficient, which is one of the parameters that can be measured in a single run, represents the dual control exerted on the growth process by mass and heat transport. It is given by:

$$\frac{1}{D_{\text{eff}}} = \frac{1}{D} + \frac{b L}{K},$$

where  $D$  is the vapor diffusion coefficient,  $L$  the latent heat of condensation,  $K$  the gas thermal conductivity, and  $b$  the slope of the vapor density equilibrium curve,  $dp/dT$ , at the temperature in question. Surface effects appear similarly:

$$l = \frac{\left(\frac{l_D}{\beta}\right) \frac{1}{D} + \left(\frac{l_a}{\alpha}\right) \frac{b L}{K}}{\frac{1}{D} + \frac{b L}{K}}$$

Here  $l_D$  and  $l_K$  are "lengths" in terms of basic, known physical constraints, and  $\beta$  and  $\alpha$  are the sticking and thermal accommodation coefficients respectively. The length,  $l$ , is deduced from the measurement provided  $D_{\text{eff}}$  is known. The growth, however, becomes insensitive to  $l$ , the larger the drop, so that "late" measurements can be used to establish  $D_{\text{eff}}$ .

The role of contaminants should obviously be investigated only after the successful completion of the above. Since their role is, generally speaking, twofold (both the growth kinetics and equilibrium position are changed), the experiment should either take on the additional task of locating the new equilibrium positions, or these should be predetermined.  $D_{\text{eff}}$  of course cannot be changed by contaminants in the drop, but  $\alpha$  and  $\beta$  can.

Measurements of the diffusion coefficient will be accomplished by observing the droplet growth or evaporation rate as the chamber is alternately expanded and compressed. The droplets can be generated and placed in the chamber

or they can be formed in the chamber directly through homogeneous nucleation. If most of the experiments are performed using externally generated drops then at least one set of experiments should be conducted on homogeneously nucleated drops as a check to insure the generation is not effecting the results.

The spread in droplet sizes should narrow which will permit easy measurement of the mean size by optical scattering. Even with a wide initial distribution, the first evaporation to less than  $0.1\mu$  will cause the distribution to be narrowed when the droplets are regrown.

The effects of surfactants can be studied by the use of gaseous additives. The droplets will acquire the surface film or gaseous scavenging of the surfactant materials.

#### T. 2. 8. 2 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	2
Size-droplet	
Type	
Pollutant	3
Pressure	2
Temperature	3
Relative humidity	3
Charge	
Rate of cooling	3
Time	
Sound	
Electric field	
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	



<u>Parameters</u>	<u>Variations</u>
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	

#### T. 2. 9 Procedure

General activity details are given below in a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restrictions with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient.

#### **T. 2. 10 Mission Timeline**

Until a more detailed knowledge of the techniques that will be used in the final version, the activity time line for the zero-g experimental procedure is the same as that developed for the terrestrial experiment.

It is possible that purge times and rates can be reduced but this can be determined only by testing of the actual equipment. The degree of automated control will effect the time required for some operations in both the zero-g and terrestrial experiments (see Figure T-4).

The primary change in the time line for the zero-g operation will be expansion of the duration of the expansion/contraction cycles to make full use of the zero-g environment.

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on all electrical components	5
• Check operational readiness of components	5
• Check and calibrate control and measuring circuits	10
• Turn on noncondensable gas supply and sump pump	.5
• Open expansion chamber ports	.5
• Purge system	10
• Load control program into control computer	(5)
• Load time lapse camera	(3)
• Close expansion chamber ports	.5
• Turn on thermal controls: conditioning chamber, humidifier, expansion chamber	1
• Adjust gas flow to humidifier	.5
• Adjust gas flow to drop generator	.5
• Turn on drop generator and pollution gases	1
• Turn on hygrometer, mass meter, size spectrometer	1
• Sample: relative humidity, drop size, mass, concentration	10
• Adjust humidifier and aerosol generator as required	(-)
• Open expansion chamber ports	.5
• Transfer monitoring to output of expansion chamber	.5
• Monitor sample: relative humidity, drop size, mass, concentration	8
• Close expansion chamber ports	
• Turn on: expansion mechanism, pressure control gas temperature, video laser, liquid water meter, drop size meter	1

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
● Let expansion chamber come to thermal equilibrium	10
● Turn off: drop generator, thermal controls for humidifier and conditioning chamber, drop size, mass and concentration meters, hygrometer	(2)
● Purge system (except expansion chamber)	(4)
● Turn off noncondensable gas supply and sump pump	(1)
● Turn on time lapse camera	.5
● Start cooling/heating program	30
● Observe chamber on video monitor	(-)
● Stop time lapse camera	.5
● Turn off: pressure control, expansion mechanism, video, laser gas temperature, liquid water meter, drop size meter	1
● Heat chamber to +10°C. Turn off expansion chamber thermal control	5
● Open expansion chamber ports	0.5
● Turn on sump pump and noncondensable gas supply	0.5
● Purge chamber	8
● Unload time lapse camera and store film	(3)
● Check computer data	(4)
● Transfer data to permanent storage	(1)
● Close expansion chamber ports	.5
● Turn off noncondensable gas supply and sump pump	.5
● Shut down system or start next experiment	10

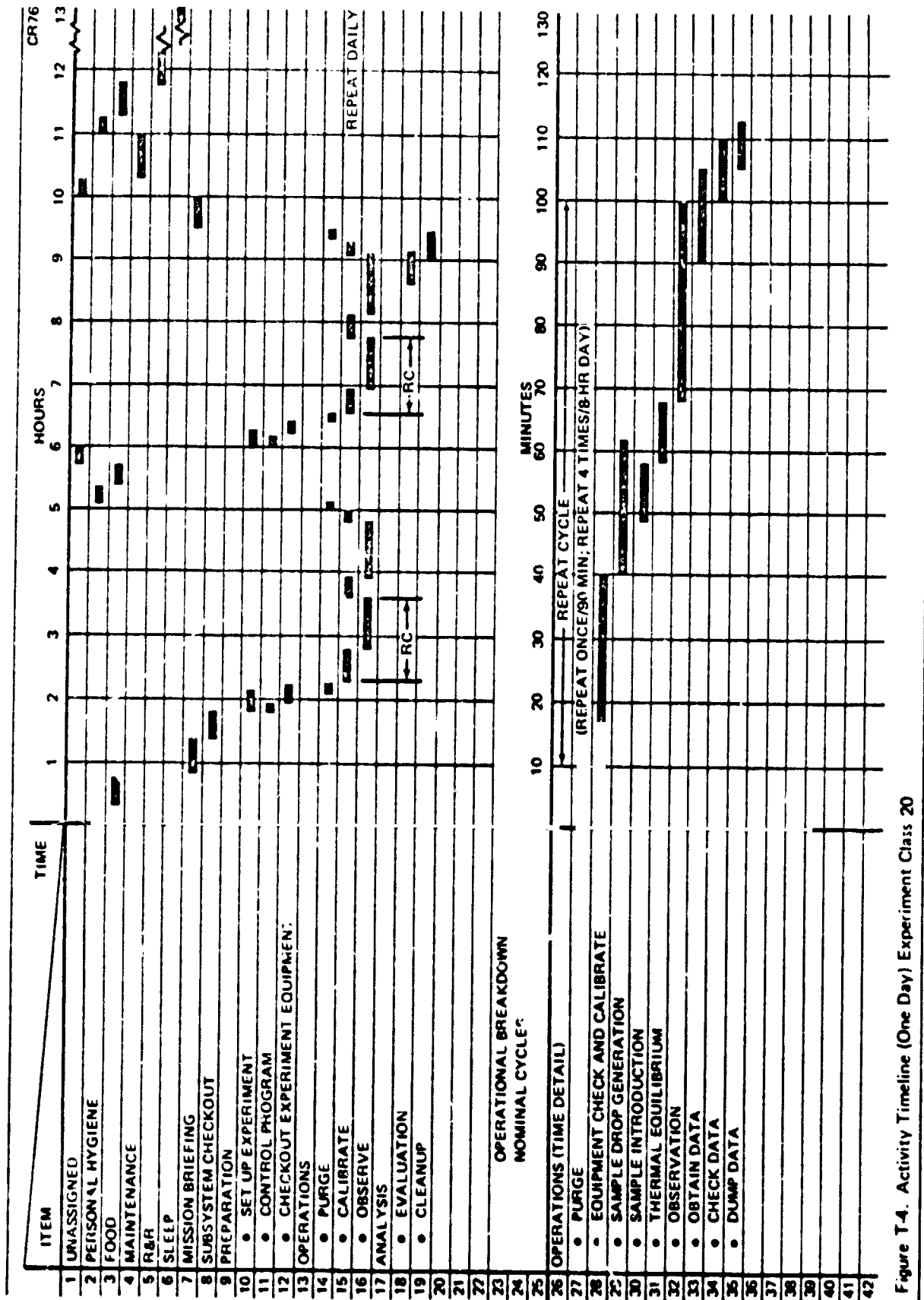


Figure T-4. Activity Timeline (One Day) Experiment Class 20

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Appendix U  
CLASS 21  
EARTH SIMULATION

U.1 INTRODUCTION

The study of the large-scale circulation of the atmospheres of the Earth, Mars, Jupiter, Saturn, as well as that of the outer region of the solar atmosphere and the earth's oceans and fluid core, consists of the study of thermally driven motion in a spherical shell of rotating fluid. Although there are intrinsic differences with regard to fluid composition and the manner in which the heating takes place, many of the observed large-scale features present in the circulations on the above bodies are thought to be related to the interaction of thermally (or buoyancy) induced motions with the latitude dependent Coriolis force. That is, rotational and gravitational forces are major constraints on the flows, but it also appears that the spherical nature of the astronomical bodies plays a very decisive role. Sphericity requires that the cross product  $\vec{\omega} \times \vec{g}$ , between  $\vec{\omega}$  the basic rotation vector and  $\vec{g}$  the gravitational acceleration vector, be a function of latitude. On theoretical grounds this latitudinal dependence seems to be crucial to the existence of equatorial accelerations. These have been observed on the Sun (Ward, 1965), on Jupiter (Chapman, 1969, among others) and have been well-documented on the Earth where they are known as the Cromwell current and the Berson westerlies. Many theories have appeared in recent years which attempt to explain some of the observations mentioned above (see Krause 1969, Busse 1970, Davies-Jones and Gilman 1970, and Durney 1970, for examples). In all theories, it is clear that the variation of Coriolis parameter with latitude is an important factor, as one would expect from the large scales of the flows being discussed.

In the past, scientists have pursued the understanding of large scale planetary circulations so necessary for effective prediction via four approaches; analytical and numerical simulation, laboratory simulation, and direct observation. Each method has its limitations. The theories contain a number of idealizations and assumptions of one form or another. The flows being modelled are three-dimensional, time-dependent, and range over many length scales; thus simplifications are necessary even in large numerical simulations using the basic equations and integrated on our most advanced computers. This situation suggests that considerable insight into the fundamental problems of global circulation might be obtained by simulation in a real physical system which retains all or almost all of the essential physics. Under normal circumstances one would think that controlled laboratory experiments would contribute. However it has been impossible to model the earth's sphericity properly in the laboratory over anything more than a few degrees in latitude. Spurious background currents are set up in experiments unless  $\vec{f}$  is parallel to  $\vec{g}$ . Since  $\vec{g}$  is unidirectional in the earth laboratory setting, many essential physical phenomena which depend on latitude variation of  $\vec{f} \times \vec{g}$  are omitted (e. g. there is no equatorial region in the classical dishpan experiments of Hide, 1958, nor are gyroscopic or planetary waves properly modelled). It is interesting to note that such mean accelerations as the equatorial jets are not observed in laboratory experiments on uniform thermal convection with rotation (Rossby, 1966) where the rotation vector is exactly aligned with gravity. The almost zero-g environment of the orbiting vehicle offers an opportunity to conduct an experiment with the necessary physical symmetries, especially a radial "gravity" field such that the  $\vec{f} \times \vec{g}$  effects are included exactly as they appear on the Earth, planets, and Sun.

Presented below is a method of generating effectively spherical geopotential surfaces by using electromagnetic forces in a particularly simple way. The resulting spherical gravity field will be seen to be comparable in magnitude to surface gravity. Thus, accurate modelling of planetary fluid flows will require that the experiment be conducted in a low g environment.



## U.2 OBJECTIVE

With controlled laboratory conditions the aspects of planetary and solar convection, with its attendant differential rotation will be simulated. The experiment will consist of a differentially heated rotating spherical annulus of fluid which possesses, in effect, a radial gravity field. The experiment, in its simplest form, is designed to incorporate the major constraints of radial gravity, rotation, and spherical geometry present in the thermal convection zones of the Sun, the Earth's core and the atmospheres of Jupiter and Saturn. The first two of these are thought to be characterized by isotropic heating. That is, the heat flux through the fluid is roughly independent of latitude and the heat flux through the fluid is roughly independent of latitude and longitude. In the laboratory, this corresponds to thermal driving which is uniform on spherical bounding surfaces. With minor changes, heating with an equator to pole differential similar to that driving the Earth's atmosphere and the oceanic thermohaline circulations can be imposed. The various regimes associated with the two types of driving will be isolated. In nature, the case of Jupiter appears particularly interesting since latitude varying heat fluxes occur from solar heating and there is also a presumably isotropic component of flux coming from the interior of the planet which is of significant magnitude.

Data will be obtained on both the structure and magnitude of the convectively driven fluid motions in the rotating spherical shell. The data will answer the following scientific questions:

- a) What is the preferred form or cell pattern of the convection subject to the above constraints of buoyancy and rotation?
- b) Is there a relationship between convective overturning and differential rotation (arising perhaps from accelerations of the equatorial regions of the "atmosphere")? Is such a relationship of the type observed in planetary and solar circulations?
- c) How does the latitude-dependent Coriolis force affect the convection? Are such affects related to the possibility of self-sustaining dynamo action?

### U.3 SCIENTIFIC JUSTIFICATION

Of the many fundamental features observed in planetary circulations, possibly the most important one to explain is the above mentioned differential rotation that seems to result from equatorial accelerations of fluid in the convection zones. Although some progress has been made in analytical and numerical approaches to the related astrophysical and geophysical fluid dynamics problems, it has not been possible to test any of the theoretical approximations because, as mentioned above, terrestrial physical experiments cannot be done except for the case where  $\vec{\tau}$  is parallel to  $\vec{g}$ . The spherical convection experiment will provide badly needed information on preferred scales and mode structure. Since many computer and analytical models rely on limited mode representations in order to obtain solutions, it is important to identify dominate modes in the real physical system and also to define the mutual and self-interactions. Progress in understanding these fundamental fluid dynamics problems will enhance the understanding of the earths' atmosphere.

In addition to being able to obtain data on the properties of solar and planetary convection, data will also be obtained on electroconvection which is important to the complicated modelling efforts and therefore related directly to the circulations on the Earth.

### U.4 APPLICATIONS

The radial convection experiment will contribute results in the following areas:

- (i) Planform selection of convection under various driving conditions when subject to latitude-dependent Coriolis forces.
- (ii) Generation of large scale mean currents by non-linear interaction of medium scale convectively driven flows. This process is of great importance in meteorology although the medium scale disturbances are generated by baroclinic rather than by convective instability. In the past local experimental models, encompassing a narrow band in mid-latitudes, have investigated some properties of non-linear or finite amplitude baroclinic waves. However no

model has previously been able to investigate the interaction of mid-latitude motions with those in the tropics or extreme polar regions. The spherical gravity experiments will be able to generate data on these interactions.

Slight modifications to the basic apparatus can yield data on the following global flow problems:

- (i) Effects of mountains on the shallow general circulation.
- (ii) Thermally driven circulations in bounded basins straddling the equator. Very little is known about interactions between circulations in different basins.

#### U.5 TERRESTRIAL LABORATORY LIMITATIONS

In a terrestrial laboratory, if  $\vec{f}$  is other than vertical, the Earth's gravitational effects will appear as an oscillatory acceleration of the stratified fluid in the rotating reference frame fixed in coordinates rotating with rate  $f$ . No such analogous forces exist in the motions we wish to model. Thus measurements of fluid flow in regions bounded by spherically heated walls contain irremovable contaminations.

An effectively radial gravity field can be established as far as thermally driven motions are concerned) by taking advantage of the temperature dependence of the polarization of the dielectric liquid in an electric field. The temperature dependent polarization force is analogous to the normal thermal buoyancy force. Both depend on temperature in exactly the same way. One would hope to make

$$R = \frac{\text{polarization force}}{\text{normal gravitational buoyancy}} = \frac{\text{"radial gravity"}}{\text{background gravity}}$$

very large. Due to material limitations (dielectric strength, for example),  $R$  cannot be made much larger than 6 in the terrestrial laboratory. However, to look at the relatively weak interactions in fluid flows relevant to solar and planetary convection one needs  $R$  to be of order 100. Thus the space laboratory with its low background accelerations is ideal for radial gravity simulation experiments. Fairly long time scales are involved in the model systems (~30 min.) so methods of producing low  $g$  environments over short time scales are not adequate.

## U.6 ZERO-GRAVITY OPPORTUNITIES

Although the solar and planetary convection experiment involves a quite distinct discipline in atmospheric science compared with cloud physics experiments, the general facility provides many of the instrumental capabilities needed to control the small spherical convection chamber. These capabilities include temperature and voltage controls and photographic data acquisition which are commonly used in many cloud physics experiments and are necessary to run the convection experiments described below. With background  $g$  levels of order  $.001$ , highly accurate and meaningful data can be obtained (the factor  $R$  relating imposed radial gravity to background noise would typically be about 200 so that we can easily observe and measure the interesting non-linear interactions with considerable confidence).

## U.7 QUANTIFICATION

The main purpose of the solar and planetary convection experiment is to test approximate analytical models of global circulation dynamics and to provide a test and input for large numerical simulations. With present (CDC 7600) computer capabilities it is not possible to integrate the governing equations with a spatial resolution sufficient to include all the active scales of motion. "Eddy viscosity" techniques are usually applied. These approximations for the small scales must be tested independently of the numerical experiments themselves. This is one area where the physical experiment is useful since it can be run in a regime which is sufficiently inviscid that a wide range of scales is present. Now it is expected that a new generation of more advanced computers will become available in the late 70's or early 80's. Nonetheless, because of the boundary layer nature of high Rayleigh number convection, many vertical grid points ( $\geq 50$ ) are needed. With global representation in the horizontal, it is anticipated that at most 10 waves in longitude may be practical (when a large number of cases are to be studied). Observations of the convective region of the solar atmosphere, for example, indicate significant energies in motions with scales orders of magnitude smaller than that contained in a ten wave model. In the proposed, over 100 waves have been observed around the circumference. Thus it is

anticipated that even more advanced computer simulations will be required to include parameterization of small scale dynamical effects. Observational (either field, or in the case of fundamental modelling, laboratory) data will be needed to verify the parameterization techniques.

## U.8 APPROACH

### U.8.1 General

We wish to replace the normal laboratory buoyancy force which is unidirectional with a radially directed force which is, like the buoyancy force in a liquid, just proportional to temperature. Now in a dielectric fluid, the body force  $\vec{F}$  has a buoyancy term arising from expansion and contraction (or change of density) with temperature, and an electromagnetic polarization force term, namely

$$\vec{F} = -1/2 E^2 \nabla \epsilon + g \rho \nabla \theta$$

where  $E$  is the imposed electric field,  $\epsilon$  the dielectric permittivity,  $\rho$  the fluid density,  $g$  the magnitude of background accelerations acting in a direction  $\nabla \theta$ . For the fluid used in our experiment (a silicone oil) the equations of state are  $\rho = \rho_0(1 - \alpha(T - T_0))$  and  $\epsilon = \epsilon_0(1 - \beta(T - T_0))$  where  $\alpha$  and  $\beta$  are constants and  $T_0$  is the ambient temperature about which experiments are run (typically 22 C) and where  $\epsilon = \epsilon_0$  and  $\rho = \rho_0$ .

Now the velocity field in an incompressible fluid (which is an accurate model for oceanic and shallow atmospheric flows – and is a primary assumption in most analytical studies of planetary circulation) with no-slip boundary conditions, is affected only by the curl of  $\vec{F}$ . This is

$$\text{curl } \vec{F} = \Delta T x(-1/2 \epsilon_0 \beta \nabla E^2 + g \rho_0 \alpha \hat{Z})$$

Both the electromagnetic and buoyancy terms are proportional to gradients in  $T$ , but the force can be directed by the geometry of the imposed electric field. The experiment takes place in the upper hemisphere of a spherical capacitor and is shown in Figure U-1. The potential difference across the electrically conducting inner and out spheres is  $V_1$  and the temperature difference is  $\Delta T$ . The inner and outer surfaces are also good thermal conductors.

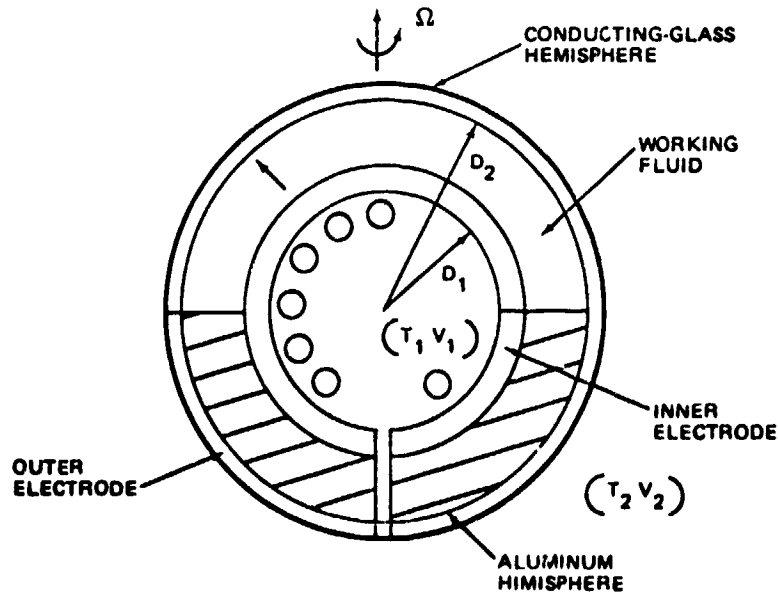
In such a bounded configuration,  $E$  depends only on  $r$  (except near the feed-through region - however, experiments run in the upper hemisphere see a field which is effectively radial as long as the gap of the annulus is not too big). It has the form

$$E = D_1 D_2 V_1 \frac{\hat{r}}{r^2 (D_2 - D_1)}$$

Thus the torque term in  $\text{curl } F$  resulting from electromagnetic effects is exactly analogous to the force resulting from buoyancy except that it is perpendicular to the radial direction. Thus we call the electromagnetically derived term a "radial gravity". One would ideally like to choose parameters such that radial gravity dominates over background gravity. The ratio of these forces is

$$2 \epsilon_0 \beta (D_1 D_2 V_1)^2 / \rho_0 g (D_2 - D_1)^2 \left( \frac{D_1 + D_2}{2} \right)^5$$

For a reasonably sized apparatus, ( $D_1$  and  $D_2$  must be chosen so that dynamical similarity is maintained with respect to the theoretical models of interest) the radial force will dominate at large enough  $V_1$ . Because of the nature of the fluids available for use in the experiments, excessive voltages are necessary to make the radial gravity field the required two orders of magnitude larger than the background gravity in the terrestrial laboratory. However in a near zero-g environment, reasonable voltages can be used with known materials to produce an essentially radial gravity field.



[TYPICAL VALUES:  $D_1 = 2 \text{ cm}$ ,  $D_2 = 3 \text{ cm}$ ,  
 $\Delta T = T_1 - T_2 = 10^\circ \text{C}$ ,  $V_1 = 2,000 \text{ VAC}$ ,  $V_2 = 0$ ]

$V_2$  THE OUTER SPHERE VOLTAGE, IS AT GROUND POTENTIAL

Figure U-1. Schematic of Spherical Convection Chamber

### U.8.2 Current Experiments

A prototype experiment has been constructed and is undergoing tests at this time. A block diagram of the experiment is shown in Figure U-2. The inner electrode of the spherical capacitor is a hollow bronze sphere. A small (10 watt) heater and a temperature sensor are located inside. The outer shell is half aluminum and half conducting glass (see Figure U-1). The working fluid is placed in the upper hemispherical annulus. The whole spherical assembly is mounted in a constant temperature bath which supplies the outer isothermal boundary condition. The outer bath and sphere is placed on a rate of turn table which is set to a fluid model rotation rate  $\Omega$ .

The data necessary to specify the dynamical regime are measured. These are the fluid properties (constant), the geometrical characteristics (constant), the temperature difference across the annulus  $\Delta T$ , the rotation rate  $\Omega$  and the voltage difference  $V_1$ . For isotropic heating these completely characterize the dynamical system through the dimensionless

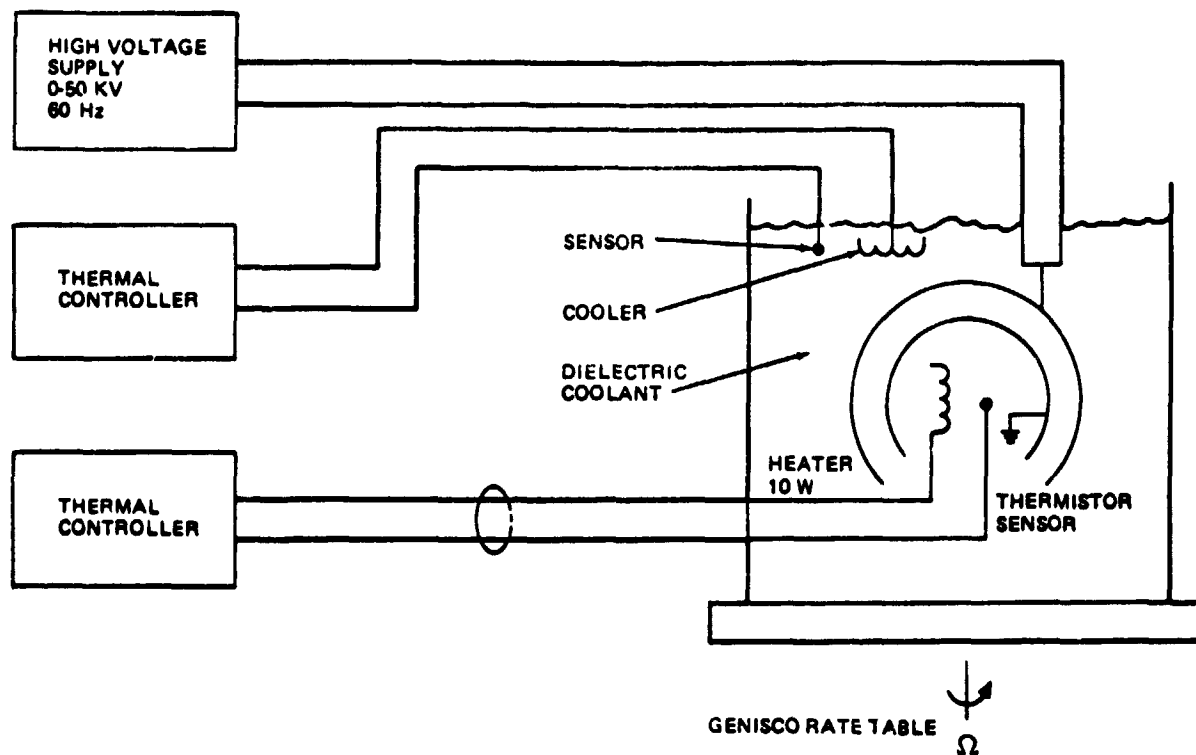


Figure U-2. Block Schematic of Terrestrial Spherical Convection Experiment Showing Control Equipment Which is Connected to the Chamber Through Slip Rings in the Turntable Base

Prandtl number	$P_r = \nu/\kappa,$
Rayleigh number	$\sim \epsilon_0 \beta \nabla E^2 (D_2 - D_1)^3 \Delta T / \kappa \nu = R_a,$
and Taylor number	$T_a = 4\Omega^2 (D_2 - D_1)^4 / \nu^2.$

There is no difficulty in getting  $R_a$  and  $T_a$  into parameter ranges which are thought (by theoretical analysis) to be significant. It would be nice to make  $P_r$  smaller ( $P_r \sim 10$  with available fluids which satisfy all the other requirements of low viscosity  $\nu = .01$  s., low dielectric loss, very high resistivity and dielectric strength). Table U-1 shows typical fluid parameters and attainable dimensionless ratios for the laboratory version of the experiment. The space laboratory version would be essentially the same.

Data corresponding to the above input parameters includes:

- 1) heat flux (e. g. power to heater) at fixed  $\Delta T$
- or 2)  $\Delta T$  for fixed heat flux
- and 3) Visualization by photography of suspended reflecting platlets which align with the fluid shear.



Table U-1  
EXPERIMENTAL PARAMETERS

---

Kinematic viscosity	$\nu = 1 \text{ cs}_1$
Thermal diffusivity	$\kappa = 6.5 \times 10^{-4} \text{ cm}^2/\text{sec}$
Ambient permittivity	$\epsilon_0 = 2.1 \times 10^{-11} \text{ (mks)}$
Thermal coef. of permittivity	$\beta = 2.5 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$
Resistivity	$r = 10^{16} \text{ } \Omega/\text{cm}$
Dielectric strength (measured in situ)	$d = 20 \text{ kv/cm}$
$R_a$ range	$< 10^3 \text{ to } 10^5$
$T_a$ range	$0 \text{ to } 10^5$

---

Most of the information obtained from the experiment comes from the streak photography\* (3, above). From such data one can recover the fluid velocity field and the much needed information covering the flow structure (characteristic wavelengths, planforms, secondary instability, etc.).

Because of lowered dielectric strength for fluids containing the necessary scatterers, the earth-based experiments are capable of maintaining a radial to background buoyancy force ratio of only about 6. Although the feasibility of such experiments has been demonstrated in the laboratory, definitive results will require much lower background g forces. Since the non-linear interactions which are thought to produce the flows peculiar to rotating spherical bodies are weak, they occur on long time scales. In the laboratory this amounts to several minutes. Thus an environment with small background g must be maintained for substantial times.

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\*See Attachment A

### U.8.3 Zero-gravity Laboratory Compatibility

Zero-gravity design of this experiment is simpler because of the lower voltages needed to attain a radial to background gravity ratio of the order of 100. The voltage needed for the apparatus of Figure U-3, with an assumed background noise of .001g is only about 3kv. This voltage level puts much less stress on the conducting glass coating, and, since it is delivered at very low current, is very safe. This potential is a factor of ten below the breakdown potential measured in actual use in our laboratory prototype. One problem that has been identified is the settle out of visualization particles in the Earth's field\*. However in the space environment with its low and fluctuating g levels, visualization particles, once stirred up, would remain suspended for the times needed to run the experiment.

In the space laboratory, a system as drawn in Figure U-1 is proposed. The outer shell is cooled (about 1 watt of heat transfer is anticipated) by a directed stream of air at a constant temperature. The chamber is mounted in a housing, free to turn on its base, which includes a synchronous drive motor and the high voltage (5 kv rms max) step-up transformer. All control circuits and sequencers are external to the apparatus and connected through slip rings in the base. The external equipment includes:

- 1) inner sphere temperature controller
- 2) ac voltage sequencer
- 3) rotation rate controller and sequencer
- 4) controller and sequencer for data acquisition camera (stationary)
- 5) manner of recording (preferably simultaneously on film along with the visual data) measured status of  $T$ ,  $V_1$ ,  $\Omega$ , and the heat flux through the spherical annulus.

---

\*There is no basic bulk force related to  $g_{\epsilon_0}$  itself corresponding to  $g_0$ , only that corresponding to  $g\epsilon T$  which is very small. The actual g loading under the applied field is substantially less than 1 g.

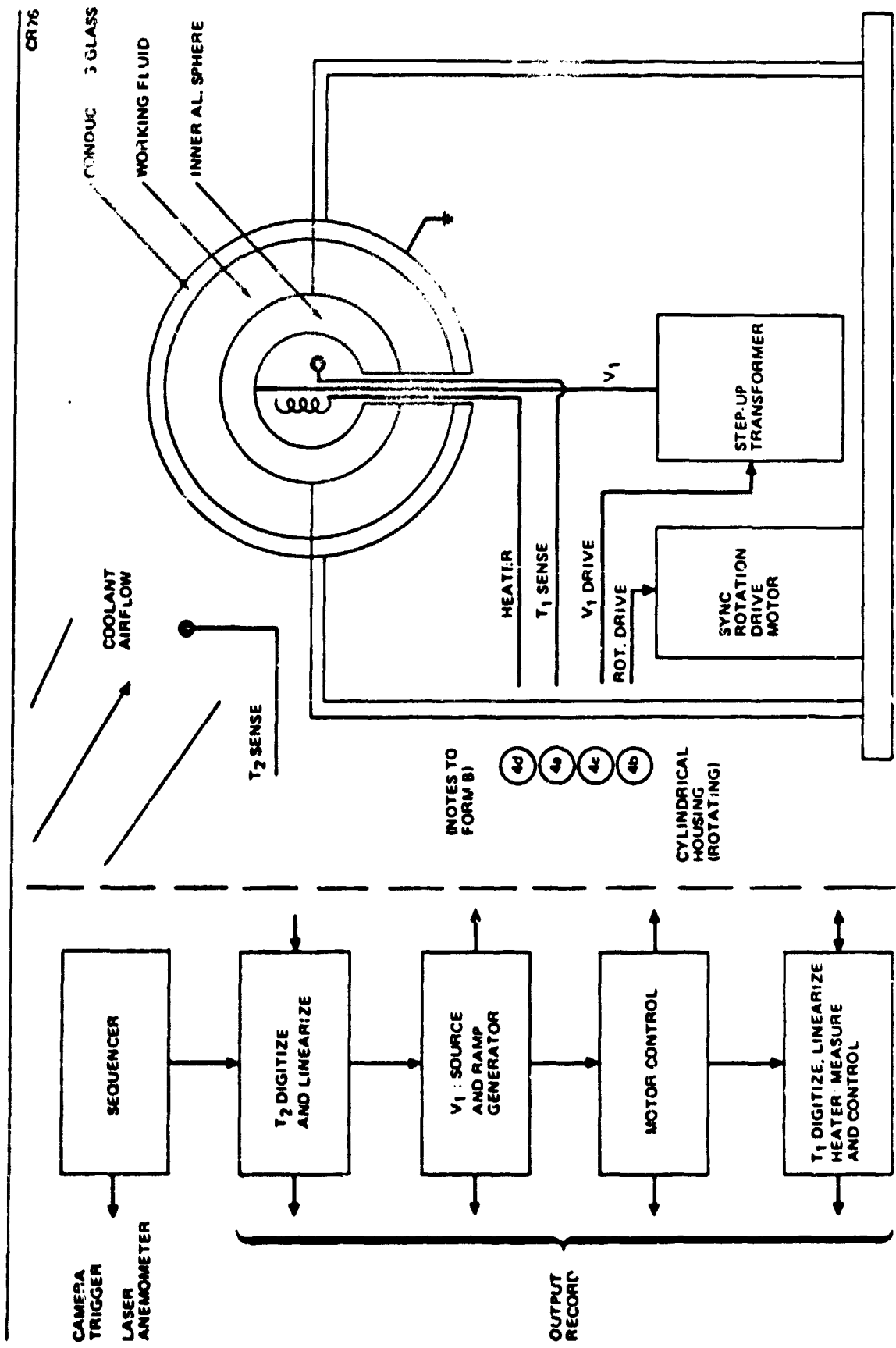


Figure U-3. Block Schematic of Space Laboratory Version of the Spherical Convection Experiment

A typical test procedure is as follows. The unit is mounted in the test area. The camera (35mm motor drive) is positioned and the power and control cables are attached to a connector on the base of the apparatus. The visualization particles are agitated for 1 minute by setting the cylinder into a back and forth mode of rotation. The first experiment is started by selecting mode 2; the cylinder is stationary (no rotation) in this mode, and  $V_1$  is automatically increased in small increments and photographs are taken every 5 minutes for 2 hours. Subsequent experiments are started by selecting mode 3; the cylinder is now set rotating with frequency and  $V_1$  is increased in small increments and photographs taken as in mode 2. Each photograph includes a field of streaks left by the particles in motion which are used to calculate the velocities and wavelengths. In addition, a readout of the power supplied to the heater and the applied voltage is included in each photograph. This is used to determine the heat flux as a function of the relevant non-dimensional parameters. Different parameter ranges are covered by repeating mode 3 with differences in  $\Delta T$  and or  $\Omega$ .

Since each apparatus is small and light, more than one may be used in order to cover cases involving boundary conditions other than constant temperature on spherical surfaces. Irregular closed basins or latitude dependent thermal input are two of the highest priority cases.

Aside from miniaturization and space-qualification of materials and construction, there is very little required development of the chamber hardware itself, as the techniques are essentially the same as those used in the terrestrial laboratory. One useful difference, if possible, would be the recovery of data on velocity vs. longitude by use of a laser doppler velocimeter. This would require considerable development, however, if laser hardware is to be part of the cloud physics laboratory, this method of data recovery could be very useful for the simulation experiments described above.

#### U.8.4 Experiment Parameters

The important parameters along with the desired variations for each parameter is given below.

<u>Parameters</u>	<u>Variations</u>
Size-nuclei	
Size-droplet	
Type	3
Pollutant	
Pressure	
Temperature	1
Relative humidity	
Charge	
Rate of cooling	
Time	
Sound	
Electric field	20
Nuclear radiation	
Adsorption	
Turbulence	
Ventilation	
Optical	
Shape	
Orientation	
Concentration	
Velocity	
Liquid-water content	
Surface tension	
Aerosol age	
History	
Ion level	
Initial conditions	
Kinetic energy	
Gases	
Spin rate	3

## U.9 PROCEDURE

General activity details are given below followed by a representative timeline. The sequence of events and indicated times are based on knowledge of terrestrial requirements and restriction with correlation of this information to a low-g environment. Additional effort will be required to make these timelines operational, effective and efficient (see Figures U-4 and U-5).

# PROCEDURE

DETAILED ACTIVITY	TIME REQUIRED MINUTES
• Turn on; temperature equilibration, load camera	10
• Inject or stir visualization particles (not needed for laser)	20
• Observation (Increase V., photograph) (measure fluxes of heat)	200 400
• Analysis (none done at time of observation)	
• Restabilize for next run	30
• Shut down, remove film	10

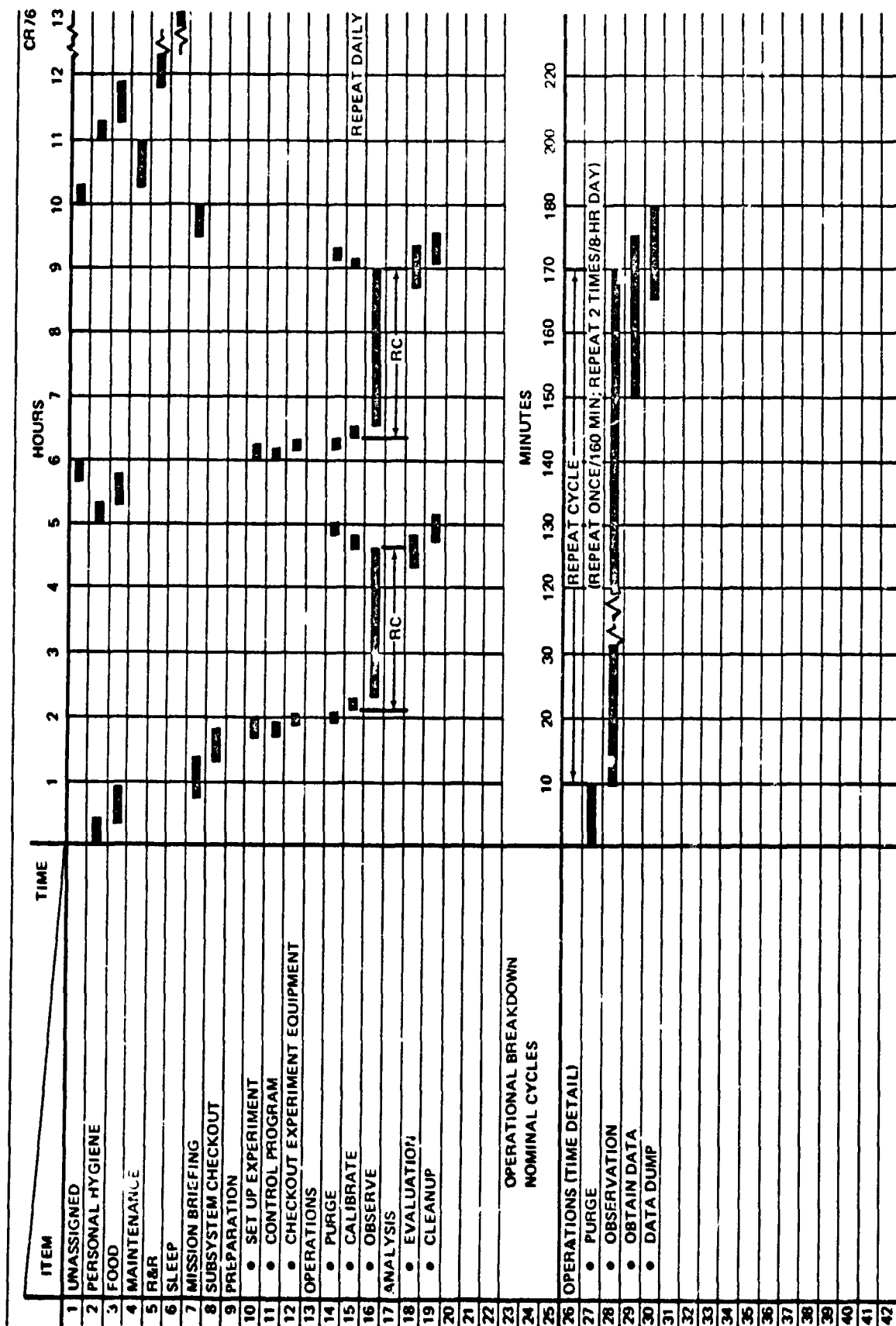


Figure U-4. Activity Timeline (One Day) Experiment Class 21



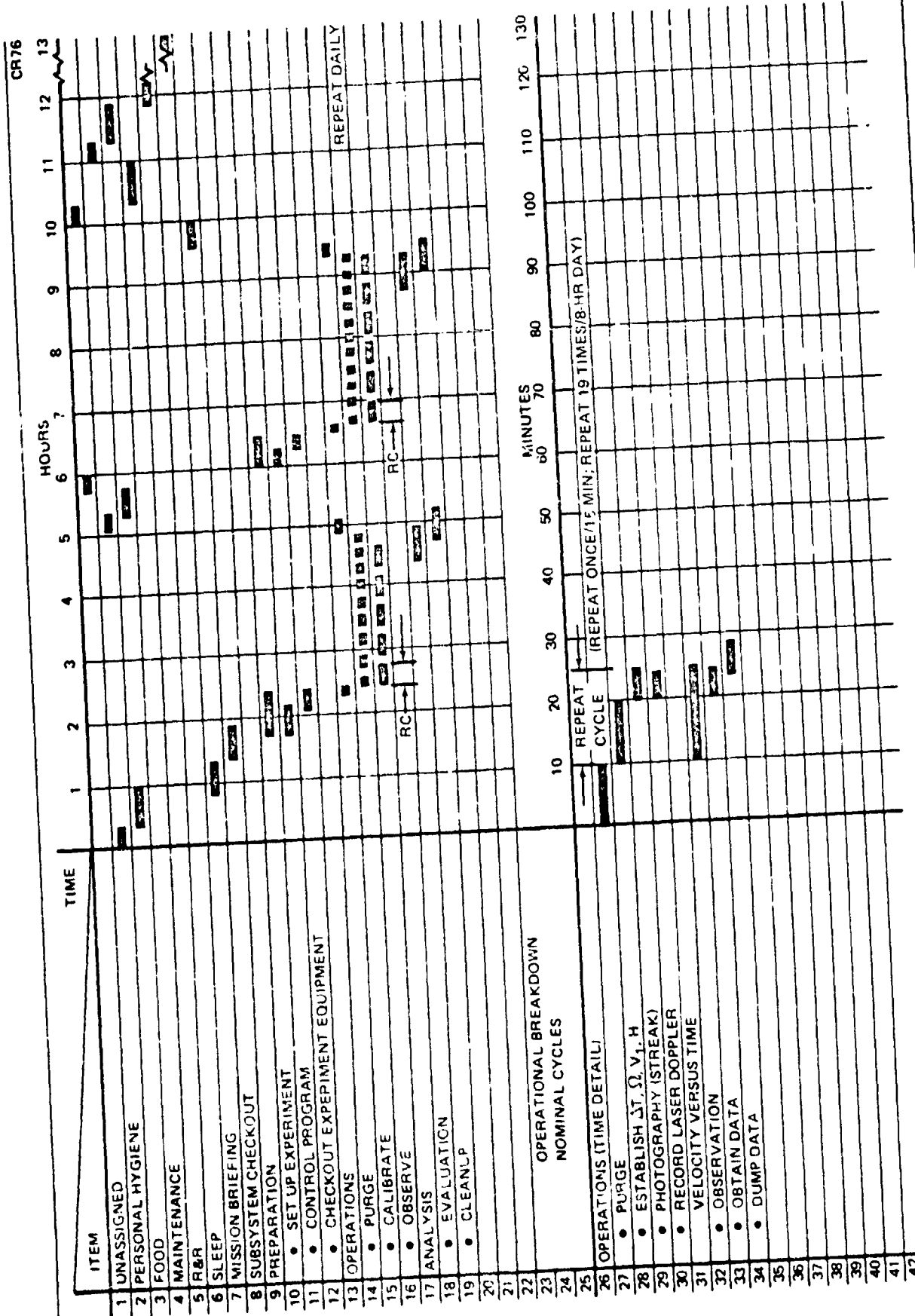


Figure U-5. Activity Timeline (One Day) Experiment Class 21

Attachment A  
LASER DOPPLER ANEMOMETRY

The streak photograph method of recovering information on the spherical convection flow field described in this section is straightforward. However the data obtained in this way only describes qualitative aspects of the flows. This data is useful and is crucial to many underlying theoretical and fundamental questions involved with thermally driven flows in the rotating spherical shell. The data obtained in this experiment is capable of resolving questions like that of the preferred planform (e. g. banana pole-to-pole rolls or axisymmetric rolls?), but will not be capable of giving detailed eddy flux measurements. Major qualitative features of the flow will be discernable but desirable details will not.

Better velocity measurement techniques should be used. In particular, the output or results from this experiment could be increased by an order of magnitude (in terms of usefulness and contribution) if a laser doppler anemometer is used to measure the fluid velocities directly. Laser anemometers are now in widespread use in terrestrial fluids labs and are commercially available (through DISA). The systems would need development for zero-g use in the convection experiment, but there would appear to be no profound difficulties involved. The laser capability should be used in this experiment and hopefully some of the technology needed could also be used in certain cloud physics experiments.

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